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rence on

# CHEMISTRY & PHYSICS OF DETONATION

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WASHINGTON, D.C.  
11-12 JANUARY 1951

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**conference on**

# **THE CHEMISTRY & PHYSICS OF DETONATION**

**MAIN NAVY BLDG.  
WASHINGTON, D.C.  
11-12 JANUARY 1951**

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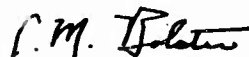
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### FOREWORD

This report of the conference on "Physics and Chemistry of Detonation" is a record of the papers presented and the discussions held at the meeting called by the Office of Naval Research on 11 and 12 January 1951. The purpose of this meeting was to bring together scientists from government, contractor, and university laboratories to discuss the current status and problems in this field of chemical physics.

It is hoped that this report will be of value to those working in the field of detonation and stimulate the interest of scientists working in related fields of chemistry, physics, and mathematics in the problems related to detonation.

To all those who actively participated in the conference, the Office of Naval Research expresses its sincere appreciation.



C.M. Bolster, RADM, USN  
Chief of Naval Research

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#### WELCOMING ADDRESS

Rear Admiral T. A. Solberg, USN  
Chief of Naval Research

Mr. Chairman, Ladies, and Gentlemen: It's always a pleasure to come to these meetings and in some respects embarrassing when you realize you have so little to contribute to the subject matter of the conference itself. Nevertheless I'm here, and it's nice to get away from a busy desk to do something like this.

I think this is the first conference in the field of physics and chemistry of detonation which has been sponsored by our office. I think it's also the first such conference that has been held since the end of World War II. One of the nice things about it is to get all the people together in this field. We haven't all been together before and perhaps it doesn't apply so much to me, but I'm sure with many of you, you see many of the faces that you probably haven't seen for a long time, or probably since the end of World War II. I think it's fortunate, too, that there are so many of the scientists here who contributed so much to Division 8 of NDRC in World War II.

As you know, the Office of Naval Research was set up under Public Law 588, and one of its duties is to coordinate research in the Navy. That is a tremendous job in itself. It is one of those things you can't force down people's throats, so you have to adopt other mechanisms. One of those mechanisms is such a conference as this one today. We've found that it has been very beneficial in various areas of urgent Naval interests—and I'm sure all of us will try to keep our efforts in urgent Naval interests—to have such symposia as this. It certainly gets the people together. It is one of the best means of disseminating information and getting exchanges of ideas. I am very much impressed with the logs of such conferences as this that I've read because of the amount of information that I see coming out of them. I learn a great deal more from reading the logs of these conferences than I could possibly get from reading a stack of reports on the same subject.

We've been trying to develop this scheme as much as possible, and about a year and a half ago we had a very successful one in another field and I felt that the method we adopted then was very important. In other words, previous to that time most of such gatherings as this had been only with the inclusion of Service personnel. At that particular one I thought it was well to bring in not only the people interested in the subject, such as our research contractors and Service people, but also to bring in the interested contractors from industry. In other words, we selected people from industry or firms whom we felt had the facilities and also the personnel to go into the development of whatever was going to come out of research in that particular subject.

One of our problems that we are striving hard to improve is the cutting down of the horrible time gaps that usually exist between the time you get some research information and you get it into use or into an end item. We felt that bringing in industry at the proper stage of the research would be a big help. As a matter of fact, in years past, I have complained because of the fact that industry doesn't allow Service people to get in on their research at some stage when we can be helpful to them in specifying the final engineering of whatever is going to come out of their research. Generally, we have to wait until we see a newspaper ad that here's something we need. Most of the time we have to go back to that particular industry and ask them to re-engineer it to suit our specifications.

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I think I would like to say just a few words with regard to our present situation and our policies under present conditions. As you know, there is a great effort being made now to produce more hardware and more materials which are available and which are the best we can get. However, I think it is very important that we also produce items which are improvements over what we have if we are going to contend with the numbers which may be against us. Consequently, it is more than ever important that we get our research information into use. I feel that in a conference of this type you should give much attention to that. Most of you know the programs; you are familiar with them; you have an opportunity to discuss them and I feel that you should start roping in those programs and the information that has been made available to you. Think in terms of what implications are coming out of this particular field of research. How can it be applied even if it's only in bits and pieces? That is something that we try to do in the Office of Naval Research. We feel very strongly that you must not wait until a particular research project is finished before you take a hard look at it; as it progresses, try to find out where implications are coming out and at least warn the people who are going to be concerned with those particular implications.

I think, also, in this conference you have an opportunity of learning more about Naval requirements, where our urgent areas of interest are, and I hope you'll prospect that also and, if possible, give off-the-cuff solutions. If you don't have off-the-cuff solutions, when you go back carry those problems with you and bring us any information that might contribute to those particular solutions.

I particularly like the way this conference is named—"Physics and Chemistry of Detonation." I can't remember many other times when I've seen two fields of science married together this way, and I feel very strongly that marriage of the sciences in almost any activity is a matter of the greatest importance. In other words, no one particular scientific subject should have a wall built around itself or be compartmented. I might even be so bold as to suggest that, knowing our own projects in this field and also having the knowledge of how a great many of our projects in the mathematical sciences are contributing and speeding up the work that is being done in this particular subject, it might also be appropriate to name this "Physics, Chemistry, and Mathematics of Detonation." I had the pleasure of getting to know John von Neuman over in London, and it was kind of hard trying to find out what he was doing. He had another mathematician with him—John Calkin—who was continually working out mathematical formulae and problems for Dr. von Neuman. He was running around seeing what damage detonation had done and developing the mathematical formulae that were involved. We all know now that he was thinking in terms of something bigger than what we had over there. He was certainly trying to develop the art as mathematically as possible as to the results of detonations, and that made me realize how important mathematics was in this whole field, whether it's in actual manufacture or in observations of the results.

I would like to add just a little bit as to how we are handling our research at the present time under the present conditions and what our policy is. In our Office we feel it is well for everyone to continue the basic research program at its present level and on its present basis. However, in connection with that, we have continually, and more fervently since Korea, been looking at what we thought were the most urgent areas and also areas which seem to be nearing completion or fruition and putting a great deal more emphasis on those, trying to speed them up. Also, with the help of our university personnel and our own Office, we have been looking very hard for implications that are coming out of the program, and in those areas where it is desirable, getting those into the applied research stage. We are also increasing our coordination and dissemination of information particularly to industry where they'll be able to pick up the ball when it is ready for them. Consequently, the extra money that we are getting we're using principally for those two activities, to put more emphasis where it is necessary and getting some things started in applied research with the idea of having the Bureaus take it over as soon as they can. Many of those projects of that type are being done on a joint basis with the various Bureaus and the other Services.

I think that is all I have to say except that I'm glad to be here and glad to welcome you all to this conference.



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**SESSION I**

**11 January 1951**

**REVIEW OF RECENT STUDIES**

**Stephen Brunauer  
Bureau of Ordnance  
Chairman**

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## RECENT STUDIES IN BUORD

Stephen Brunauer  
Bureau of Ordnance

I feel very good about this meeting. I feel almost as though I were present at a class reunion to see some of the faces of the old friends with whom we worked together in World War II. At the same time I'm sorry for two things. First of all, the occasion that brought us together is not a joyful one. The peace that followed the war in which we worked together apparently was too short-lived, and we are again in a national emergency, and the chances seem to be awfully slim that we can get out of another world war which would be the third in the lifetime of most of us.

The second reason I feel sorry is that some of our friends are not here, some of our old collaborators like John von Neumann, E. Bright Wilson, MacDougall, Eyring, Gamow, and others. I know, however, that these men did not lose their interest completely in explosives. In fact, some of them are working full time in the field, and I do hope that when the next gathering comes, they will all be here, and in between they have helped us also in our work.

There is another reason why I feel good about this meeting, besides seeing the old faces. I see many new faces here. It is to the credit of the Services—and especially of the Office of Naval Research, the Ordnance Corps of the Department of the Army, and the Bureau of Ordnance of the Navy Department—that they kept up the interest in this field through the past years and frequently under rather adverse circumstances.

Now as far as the purpose of this meeting is concerned, I can not add much to Admiral Solberg's presentation. He gave it in more detail, but perhaps I'll repeat one or two points about the purpose of this gathering. The ultimate purpose as I see it - and I use the word "ultimate" as contrasted to the immediate purpose - is to improve the weapons of the Services of the United States and to improve them by the particular method that we are all interested in, namely, by explosive means. That improvement may be one of two types: It may be by putting new and better explosives in our weapons, and the other is by controlling the explosion process in such manner that we can get greater effectiveness out of our weapons. The immediate purpose of this gathering is to increase our fundamental understanding of the detonation process. I do not need to show to any of you that there is a very close connection between the ultimate and the immediate purpose, namely, that the increase in our understanding of the detonation process would in the long run, or perhaps not-so-long run, bring about improvements in our weapons by explosive means.

Now there are two ways in which this conference can help increase our fundamental understanding, as was pointed out by Admiral Solberg. One is by giving us an opportunity here to exchange ideas with each other; and the second, which perhaps is even more important, is that it gives you an opportunity to get acquainted with each other and to get acquainted with each other's problems. This conference will last only two days, so you can exchange ideas here for only two days, and perhaps we may not have another one for another year. But in the intervening period, if you know each other and each other's problems, you can keep on exchanging ideas among yourselves and thereby help each other in increasing our knowledge of the detonation process.

Those of us who worked together in World War II know it full well, and those of you who are newcomers in the field probably realize it too, although perhaps to a lesser extent, what

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a tremendous contribution was made to explosive research by the increase in our understanding of the explosion process that was brought about by the work of men like Kistiakowsky, Kirkwood, Wilson, von Neumann, MacDougall, Brinkley, Richardson, Eyring, Gamow, Cook, and many others. The ideas that they developed, the understanding that they brought about, was a tremendous help to the experimentalists, and together the theorist and the experimentalist brought about very important improvements in the weapons of all three Services—or rather at that time there were only two Services of the United States—by explosive means.

At the end of World War II our theoretical understanding was almost adequate to explain the process, the actions of explosives that were used in that war. To be sure, it was realized that the theory was not entirely perfect. Certain limitations were known. It was believed, for example, that perhaps a better equation of state might be found, or that the evaluation of the parameters in the equation of state, especially the covolume factors, may be improved. However, now on the basis of the work that has been done since that time, we realize that the theory might have even greater limitations. We've been working since that time with explosives of the type we did not use in World War II. For example, when the Naval Ordnance Laboratory investigated the detonation velocity of the compound hydrazine mononitrate, a compound that contains no carbon, they discovered that the theoretical value of detonation of velocity was around 6000 meters per second, but the velocity actually measured was around 8000 meters per second. Other differences came in when we dealt with explosives that had much higher percentage of solid products than the explosives we used in World War II, which perhaps would require some modification or at least a more detailed understanding of the detonation process. Then difficulties came up also in the study of underwater-explosion phenomena. The Kirkwood theory gave very well the parameters of the shock wave, especially under water, but in air too, for action beyond several charge-diameter distances, but it isn't quite as good if we deal with the parameters of the shock wave closer to the explosion. What is more, only recently did we realize the great importance that the bubble produced in an underwater explosion plays in damage to underwater targets, and in order to evaluate the bubble parameters we probably need again a more detailed knowledge of the detonation process than we needed for the evaluation of the shock-wave parameters at greater distances.

Now, to say just a few words about the efforts since World War II. The situation was this: in the first two or three years following the war we were building up research. We were building up personnel to do the research and facilities and instrumentation for further research. So these two or three years were very meager in terms of results that you could use in your discussion today. Then in the last two years we were having economic and financial difficulties. You know about that—all of you—and these difficulties hit rather hard at explosive research and especially the phases of explosive research that will be discussed here, the fundamental phases of research. However, in spite of all of that, certain new things did come out and you'll hear those at this meeting.

In conclusion I would like to thank you together with Admiral Solberg for appearing at this meeting and giving your time to it, and I wish you a very pleasant and fruitful conference.

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## RECENT WORK ON DETONATION AT ABERDEEN

Jane Dewey  
Terminal Ballistic Laboratory  
Aberdeen Proving Ground

### MEASUREMENT OF DETONATION VELOCITY

One completed project(1) includes a study of errors in determining detonation velocities. In investigating detonation velocity in Primacord, it was found impossible to obtain the expected reproducibility. Fourteen measurements were made on the same strips using a rotating-mirror camera and 1.6-megacycle counter chronographs simultaneously. Although the precision of a measurement as estimated by comparison of the two methods is about 2%, the standard deviation of camera measurement estimated from the 26 rounds fired is 6%. The range of the measurements by the camera and electronic methods is 15 and 9%, respectively, in the 14 rounds studied by both methods. The difference between the standard deviations of the camera and electronic methods is insignificant. Both make it clear that the Dautriche method may introduce a 10-20% measurement error.

### AIR SHOCK CLOSE TO CHARGES

Ultra-high-speed photographic measurements(2) (exposure time about  $3 \times 10^{-8}$  sec) on air shock close to spherical Pentolite charges have progressed to the point where a tentative report can be given. As in previous Bruceton(3) data, a linear decay is found close to the charge. All peak-pressure data obtained in this laboratory on these charges are represented to about 10% by the three-parameter formula

$$PZ = [2.18 + 71.4/(1 + 0.23 \text{ lb}^{2/3} \text{ ft}^{-2} Z^2)] \text{ atm.ft/lb}^{1/3},$$

where P is the peak pressure and Z the scaled distance. The range of Z's is from the charge surface ( $0.133 \text{ ft/lb}^{1/3}$ ) to  $100 \text{ ft/lb}^{1/3}$ . Gage as well as velocity measurements cover the range of pressures from 25 to 0.015 atmospheres. Further data at the high and very low pressure ranges are required. The lower portion of the curve gives a good representation of the data of other workers; especial attention was given to the measurements of Stoner and Bleakney(4).

### INITIATION BY AN EXPLODING WIRE

For blast work close to small spherical charges, spherically symmetric initiation is almost essential. This has been obtained by using an exploding wire surrounded by handpacked PETN. Desensitized PETN from Primacord detonates as readily as the pure material, but a larger quantity is required to initiate Pentolite. In most of the work one gram of PETN from Primacord was tamped by hand into one-half-inch-diameter hemispherical cavities in matching pieces of Pentolite. Using the pulse from a 0.4-microfarad condenser and #40 Nichrome wire, initiation is obtained with 4 KV. Experimental studies with a circuit of 0.15 microfarad capacity and 0.06 microhenry inductance have shown that the resistance of this type of wire must be above 0.7 ohm to initiate at any voltage. Below 5.7 KV initiation is not obtained. As the resistance of the wire is increased above the minimum value, the voltage required to initiate increases rather slowly when the resistance is near the minimum or the critical damping resistance and very rapidly at other resistances. This work is continuing with the aim of investigating its value for study of the mechanism of initiation in solid explosives.

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3. Boggs, E. M., and Hurwitz, M. D., "Optical Studies of Strong Shock Waves," OSRD 5613 (1945)
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## DISCUSSION

DR. KISTIAKOWSKY: I would like to make a couple of short remarks. In about '44 rather extensive work was done on the Primacord velocities which for various reasons cannot be put in the form of a report. It was indeed found that the commercial Primacord as supplied by the Bickford Company did show a substantial variation. However, it was also found that by asking the Company to be a little more careful—and apparently not very much more careful since the price wasn't very much higher—it was possible to get Primacord which had very much better reproducibility than the commercial. After six years I don't remember what the actual figures were, but it was an improvement by a rather substantial factor in the magnitude of the random errors. Also, it was shown that the cord coming from a given spool within itself was very high in reproducibility. The variations from spool to spool were somewhat large. Then as to the PETN detonation by an explosive wire, it is a very good method to create spherical waves, but I have a suspicion that if you are interested in determining the minimum energy required to set off detonation, you'll have to produce extremely rigid chemical controls in the crystal habitats of the PETN.

DR. DEWEY: As to the Primacord, I'm sorry to say this is Primacord Bickford claims is the best that can be made and these jumps occurred within a roll. We do not expect to experiment directly on the sensitivity of the PETN but on the sensitivity of other explosives to an entire initiator set up by this method in which you can get the maximum control of the form of the detonation wave and know the pressure in the shock emerging from the initiator.

Those measurements have been published. Camera and electronic measurements on the same piece of Primacord check in every case very closely, but both of them change suddenly after nine or ten pieces of measurements. I think you'll see why we do not think it is an optical illusion.

DR. LEWIS: We have had better luck measuring Primacord. I believe we obtain reproducibilities to within half of one percent by the methods we use, and I might also suggest that you contact the Austin Powder Company because they have been recently making Primacord which is of a rather good quality.

I want to mention very briefly what we are starting to do in initiation from point sources and measurements of minimum energy for ignition. There are two methods: one, to insert a bubble of controlled size with the explosive and initiate by ultrasonic waves of known impulse. The other method is to insert a steel ball or other metal ball into a solid explosive or liquid explosive and to initiate by very short induction heating. That work has not progressed far enough yet to give any results but I believe it has some value.

DR. HARTMAN: I suppose I should add my voice to those who say they found no difficulty with Primacord, but I had a slightly different question. I understood you to say something about the pressure varying linearly in an air shock.

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DR. DEWEY: Close to the charge.

DR. HARTMAN: And this is calculated from the velocities?

DR. DEWEY: Yes.

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STUDIES ON DETONATION PHENOMENA<sup>1</sup>

F. C. Gibson and C. M. Mason  
Bureau of Mines  
Pittsburgh, Pennsylvania

## INTRODUCTION

The classical theory of the detonation wave applies to steady nondimensional propagation in which the chemical reaction in the detonation wave is sufficiently rapid so as not to affect the detonation velocity. This treatment provides a means of evaluating the parameter of an equation of state for the gaseous products of detonation, using experimentally determined detonation velocities. Information on the thermodynamic state of the gases is necessary to detailed study of detonation processes, as the detonation state is inaccessible by other experimental means. A systematic study of detonation velocities provides a comparatively simple means of estimating the properties of the detonation gases. To obtain high precision, it was necessary to study and develop methods of preparing suitable explosive charges of uniform quality and high purity. A versatile oscillographic method has been developed for precise determination of detonation velocities.

Although detonation velocity data are sufficient for the determination of the parameters of an equation of state of assumed form, it has been shown that the information is insufficient to determine whether or not the form assumed is adequate for the description of the thermodynamic state of the detonation gases. Since the direct experimental determination of the temperature of the detonation wave would provide criteria for selecting a proper form of the equation of state, a method has been developed for determining this quantity by optical means. Although the method appears to be promising, additional refinements of the experimental arrangement will be required to permit accurate determination of the temperature of the detonation wave.

## MEASUREMENT OF DETONATION VELOCITY

The oscillographic method for determining velocity of detonation in explosives consists of an electronic system which produces a steplike calibrated trace on the cathode-ray tube of an oscillograph that is recorded by a simple photographic process. Figure 1 shows a block diagram of the apparatus. The impulse pickup through the explosive stick or cartridge depends on the change in resistance at the pickup station due to the highly ionic flame front passing the wires that comprise each make station.

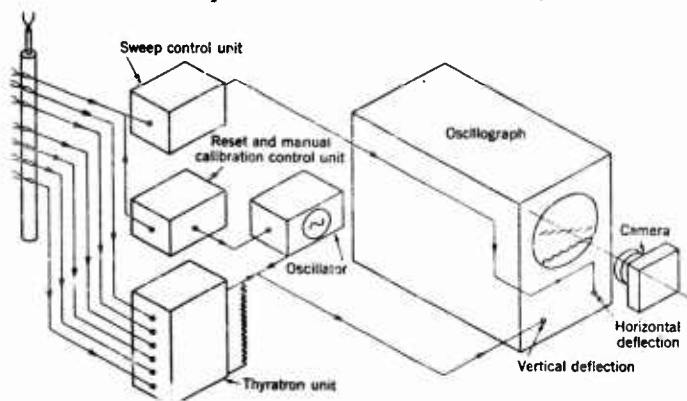


Figure 1 - Block diagram of the apparatus

A potential of approximately 100 volts exists between the two wires that comprise each station; the wires have been twisted and introduced through the explosive charge and are insulated from each other only by the enamel covering. When the front reaches a station, a sharp potential rise results, causing the thyatron associated with the station to trigger or conduct. This is a rapid phenomenon, requiring approximately 0.05 microsecond for conduction. Each thyatron station is influenced by the identical stimuli from the explosive

<sup>1</sup>The detonation luminosity and temperature measurements described in this report were carried out by R. L. Boyer.



charge so that, regardless of the position of the ionic cloud with respect to the detonation front, a true measure of detonation velocity is obtained. Thyratrons were chosen because a thyatron responds only to an initial pulse and is completely stable until the end of the test, at which time it is reset manually. The unit now in use at the Bureau of Mines Explosive Testing Station consists of five channels for determining average velocities of detonation across five adjacent short segments of an explosive charge. The unit employs seven thyatron tubes, a crystal-controlled oscillator, a standard laboratory high-speed oscillograph with a suitable recording camera, and the necessary associated power supplies. One thyatron is used for synchronization purposes and the remaining six for velocity determinations.

Since a driven sweep is employed, it is necessary to synchronize the sweep with the explosion so that the phenomena to be measured fall on the trace and can be photographed. Consequently, the sweep is started a few microseconds before the timing interval begins by a synchronization "make" station a few centimeters ahead of the first measuring station. The six thyratrons have a common resistor in their plate circuits. An increase in voltage drop appears across this resistor, which is between the vertical deflection plates of the cathode-ray tube, as each thyatron conducts. Consequently, as the beam spot progresses across the face of the cathode-ray tube, it is deflected vertically as each station is reached by the flame front and the corresponding thyatron conducts. A steplike trace appears on the tube and is recorded by means of a still camera whose shutter has been opened for a time exposure just before the charge is fired. As soon as the explosion phenomenon has been recorded, a calibration time base is superimposed on the record by means of a 1-megacycle, crystal-controlled oscillator. This time base is highly reliable, since the quartz-crystal frequency is very stable and its resonant frequency can be compared with the standard frequency transmissions of Station WWV. This comparison is seldom necessary, since low-drift, temperature-compensated quartz crystals are available. The record is analyzed by means of a standard 10-power laboratory microscope, utilizing a cross-hair eyepiece.

An actual velocity record is shown in Figure 2. The charge was prepared of 20-micron sized PETN. Each step interval represents the time required for the detonation to traverse 3 cm. The charge was prepared at three densities - 1.51, 1.21, and 0.90 gm/cm<sup>3</sup>—with two segments of each and initiated from the high-density end. One step is shown for the highest-density segment and two for each of the lower-density segments. The velocities obtained for the composite are within 1.5 percent of those predicted by the linear relation between the velocity of detonation and the loading density, established by data obtained previously by several methods. The increase in the step length is easily discernible on the record by unaided visual inspection. The time between adjacent peaks of the calibration wave is 1 microsecond.

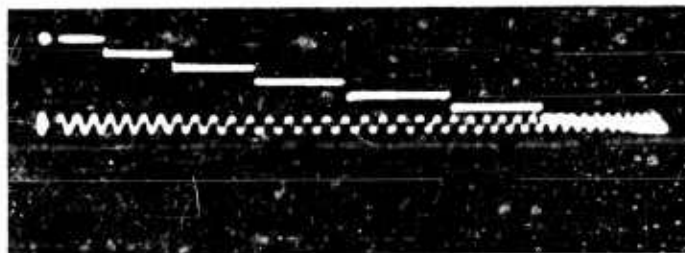


Figure 2 - Typical cathode-ray oscillograph trace for determining velocity of detonation across five adjacent segments of a three-density PETN charge

The apparatus has been thoroughly tested by measurement of average velocities of detonation over 10-cm lengths on solid explosive charges between two stations. Standard explosives were used, the early work being performed on Primacord. Typical values for charges prepared from adjacent segments of Primacord are shown in Table 1. The maximum deviation from the average for four charges is 0.5 percent. Typical data for cast charges of (50/50) Pentolite are also given, in which the maximum deviation from the average is less than 1 percent for six charges that were homogeneous and uniform in quality.



TABLE 1

## Velocity of Detonation for Primacord\*

Charge No.	Station Distance, cm	Time, Microseconds	Velocity of Detonation, m/sec
1	10.0	15.5	6450
2	10.0	15.5	6450
3	10.0	15.5	6450
4	10.0	15.6	6410
			Average 6440
Maximum deviation from average, 0.5 percent			

Velocity of Detonation for Pentolite (50 Percent TNT - 50 Percent PETN)			
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Charge No.	Density	Station Distance, cm	Time, Microseconds	Velocity of Detonation m/sec
1	1.69	10.0	13.30	7520
2	1.70	10.0	13.40	7465
3	1.70	10.0	13.40	7465
4	1.69	10.0	13.45	7435
5	1.69	10.0	13.35	7490
6	1.67	10.0	13.50	7405
				Average 7465
Maximum deviation from average, 0.8 percent				

\*The deviations of these data would be further reduced if calculated to a common density by means of an appropriate  $D$  vs.  $\rho_0$  law.

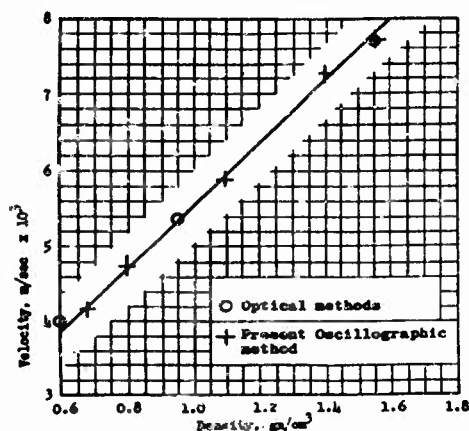


Figure 3 - Velocity of detonation for PETN of approximately 30-micron particle size

The reliability of the method is further illustrated in Figure 3, where velocities of detonation at five loading densities are compared with data accumulated by many tests, employing optical methods. The oscillographic rates were determined over 10-cm distances, and the average of two tests at each density is plotted. The variation is of the order of 1 percent.

A program has recently been initiated to study the decay of nonsteady detonation waves as affected by changes in particle size and loading density of TNT charges. Previously obtained data are not accurate enough for this purpose, much of them having been obtained as averages over segments of considerable length. Charges were prepared of two particle sizes, 500 and 1000 microns, at three loading densities. The charges were 3/4 inch in diameter and confined with a single wrap of cellulose acetate sheet. Each charge was amply initiated by a tetryl booster followed by a

high-density pressed TNT pellet. In Figure 4, the average velocity over 2-cm segments of charge is plotted against distance from the high-density TNT booster. In the case of 500-micron sized material, velocities for the three loading densities, 1.28, 1.15 and 0.95 gm/cm<sup>3</sup>, tend to a stationary value in a distance of 7 cm or less. In the case of 1000-micron sized material, a stationary value is obtained for a loading density of 1.27 gm/cm<sup>3</sup>; however, for densities of 1.13 and 0.95 gm/cm<sup>3</sup> the velocities do not attain stationary values over the distances investigated, and in the case of the lower density the detonation wave has nearly died out in a distance of 5 cm.

This instrument will permit study of the variation of detonation velocity between adjacent charge segments of unequal density, unequal charge diameter, and unlike materials. Since the velocity can be accurately determined over relatively short segments of charge, a comparatively small amount of material is required for each measurement. This constitutes a particularly attractive feature of the instrument, since preparation of charge material of high purity and controlled particle size is usually difficult and tedious.

#### MEASUREMENT OF DETONATION LUMINOSITY

The parameters of various forms of the equation of state for the gaseous products resulting from the detonation of solid explosives have been evaluated in recent years by Brinkley and Wilson,<sup>2</sup> Caldirola,<sup>3</sup> and Cook.<sup>4</sup> These calculations have been based on the classical hydrodynamic theory of detonation, together with thermochemical data and experimental values for the detonation velocity of high explosives at various loading densities. These equations of state permit the calculation of the pressure and temperature of the detonation wave. Pressures as high as 200,000 atmospheres for the detonation wave at high loading densities of explosives are indicated by the different equations of state. However, calculations of the detonation temperature vary between 3500° and 6500° K, depending upon the particular form of equation selected. The considerable variation of the detonation temperature as calculated by the several equations of state indicates that an experimental measurement of the detonation temperature at several loading densities would be useful to determine the proper form of the equation of state and provide approximate values for its parameters.

A radiation method appears to be most appropriate for measuring the high temperatures of the detonation wave. A method of this type has been developed in this laboratory, and preliminary tests of its application to the determination of detonation temperature have been carried out. The method is based on the change in spectral distribution and the density of the radiation energy as the temperature of a black body increases. Values of the radiation energy density at wavelengths between 0.30 and 1.50 microns for black body temperatures of 3000°, 4000°, 5000°, and 6000° K are shown in Figure 5. These values may be calculated directly from Planck's formula<sup>5</sup> for black body radiation and have also appeared in tabular form<sup>6</sup> for several temperature ranges.

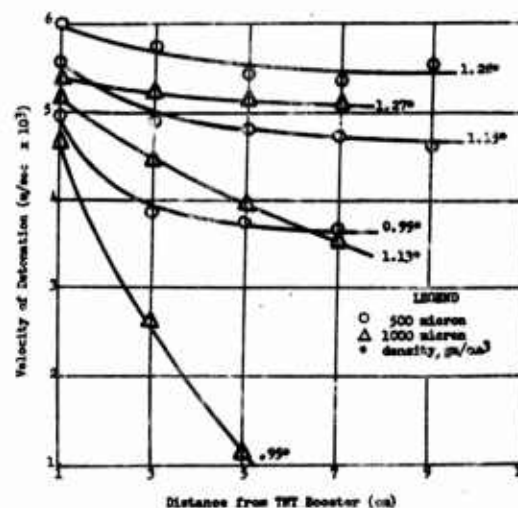


Figure 4 - Velocity of detonation decay for 500- and 100-micron particle sized TNT at three loading densities

<sup>2</sup>Brinkley, S. R., Jr., and Wilson, E. B., Jr., OSRD Report No. 905 (1942)

<sup>3</sup>Caldirola, P., J. Chem. Phys., 14, 738 (1946)

<sup>4</sup>Cook, M. A., J. Chem. Phys., 15, 518 (1947); 16, 1081-1086 (1948)

<sup>5</sup>Planck, M., Ann. Physik 4, 553 (1901)

<sup>6</sup>Smithsonian Physical Tables, 8th ed., The Smithsonian Institution, Washington, D. C. (1934), Tables 309-311, pp. 314-316

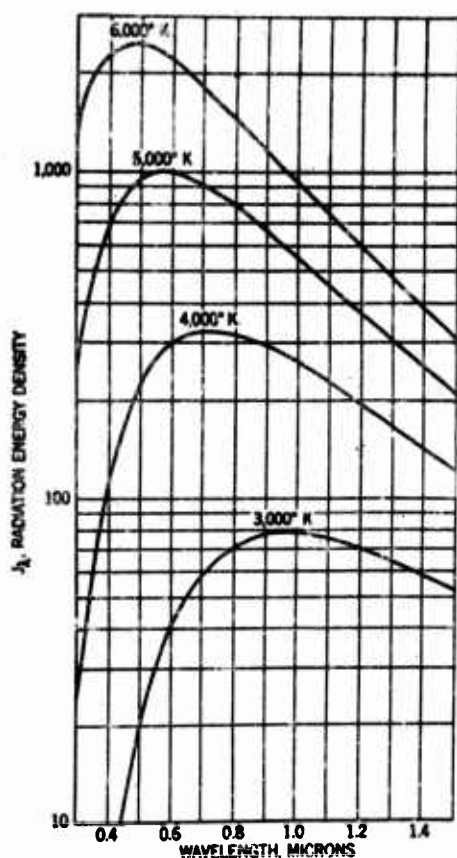


Figure 5 - Black body spectrum intensities

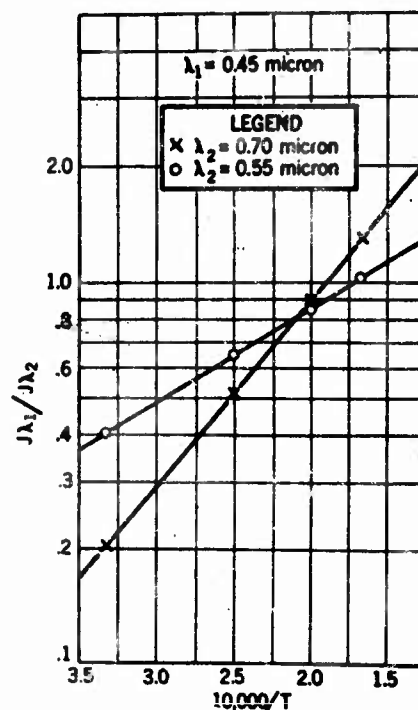


Figure 6 - Relative radiation intensities  $\lambda_1 = 0.45$  micron

The relative intensities of the radiant energy at two wavelengths,  $\lambda_1$  and  $\lambda_2$ , for each of the temperatures are determined from the values of radiation energy density. For example, at  $3000^\circ\text{K}$ , the ratio of radiant energy for  $\lambda_1 = 0.45$  micron to that for  $\lambda_2 = 0.70$  micron is 0.206. As the temperature increases, this ratio increases, and at  $6000^\circ\text{K}$  the relative intensity of the two wavelengths is 1.32. This change in relative intensity with black body temperature is shown in Figure 6 for two wavelength ratios, and a linear calibration is obtained when the logarithm of the intensity ratio is plotted as a function of reciprocal temperature. The line with the steeper slope shows the relative intensity of radiation for wavelengths of  $\lambda_1 = 0.45$  micron and  $\lambda_2 = 0.70$  micron, while the second line represents this ratio for the same value of  $\lambda_1$  but  $\lambda_2 = 0.55$  micron. This type of calibration line can be used in the experimental measurement of temperature, except that it is preferable to base the calibration on measurements of the radiation emitted at selected wavelengths by radiation sources at various temperatures.

Spectral zones of narrow range of wavelength are obtained with interference-type optical filters, and the transmitted radiation is detected by multiplier-type phototubes and amplifier networks. Incandescent lamps were used as radiation sources between  $3000^\circ$  and  $3450^\circ\text{K}$ ; their temperatures were determined by operation of the projection-type filaments at voltages

between 85 and 115 and the direct observation of the filament temperature with a potentiometer-type optical pyrometer. A xenon-filled, high-intensity flash tube with an estimated color temperature of 6800° K was also used as a calibration point. Experimental calibration data for these radiation sources are shown in Figure 7 for optical filters with maximum transmissions at 4510 Å, 5420 Å, 6420 Å, and 6870 Å; two additional calibration lines are obtained by using the relative intensity ratio for wavelengths 4510 Å and 6420 Å and for wavelengths 5420 Å and 6870 Å. The use of four filters with transmission in various parts of the visible spectrum permits four observations of the radiation temperature for each measurement.

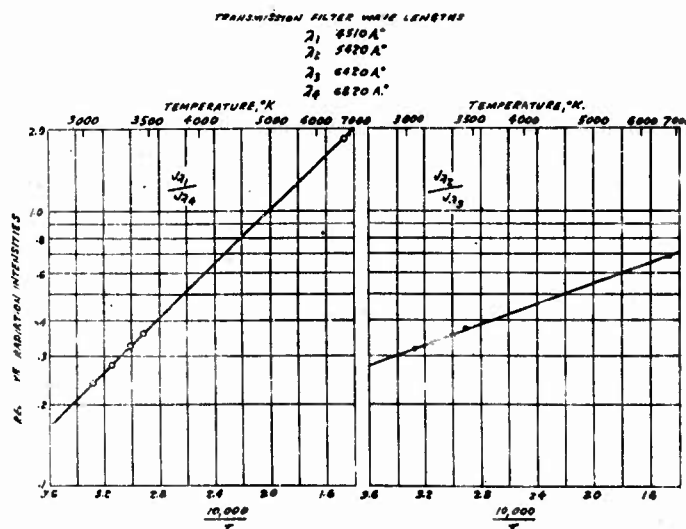


Figure 7 - Experimental calibration data for radiation-temperature measurements

The schematic diagram of the proposed apparatus for measuring detonation temperatures is shown in Figure 8. The source of the radiant energy X corresponds to the lamps for the calibration data and to the explosive for determining the detonation temperatures. This source is placed in an enclosed bombproof approximately 1 meter from the optical filters and phototube detectors. Two "multiple" glass windows, each of 1-inch thickness, are used as transparent apertures through the bombproof wall. A steel tube with slit aperture near the radiation source protects one of these glass windows and also limits the effective radiation zone of the explosive that is incident upon the filters and phototubes. An insert is used with this steel tube to obtain narrow slit apertures; black masking tape over these inserts permits slit aperture widths down to 0.8 mm in the direction of the detonation wave along the length of the explosive charge. The explosive charges are set approximately 8 cm from this slit aperture, and plastic windows of 1-cm thickness are cemented over the aperture to prevent fracturing of the glass window by the exploding charge.

The luminosity from the explosive passing through the slit aperture is incident upon a set of four multiplier phototubes behind the glass window in the bombproof wall. Radiation intensities for the different regions of the visible spectrum are obtained by placing the narrow-band optical filters directly in front of the sensitive surfaces of the phototubes. Each of the phototubes is connected through a vacuum-tube amplifier to the vertical plates of a cathode-ray tube

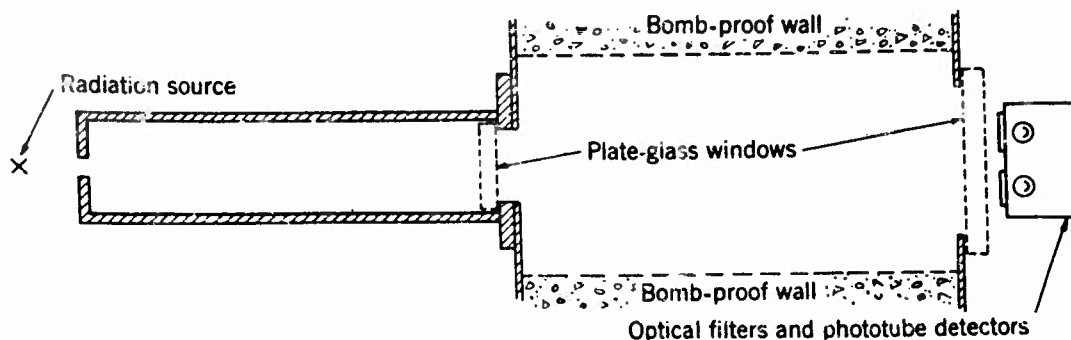


Figure 8 - Schematic diagram of optical system

so that luminosity-time records at four different wavelengths of radiation are obtained on the driven sweep of a four-channel oscilloscope. These four luminosity-time curves for the detonating explosive charge are photographed on a single frame of 35-mm film with a miniature-type camera and f:2.0 lens. Peak deflections on the luminosity-time curves are measured directly from the film negative with a traveling microscope, and the peak intensities are evaluated as equivalent currents in the phototube circuits by comparison with the experimental calibration of the current sensitivity of each phototube-amplifier network. Detonation temperatures can thus be determined for each explosive charge from the ratios of the equivalent phototube currents at the different wavelengths employing the experimental calibration curves.

Time synchronization between the detonation wave advancing along the explosive charge and the triggering of the sweep trace of the oscilloscope is obtained by an electrical circuit similar to that used in the velocity of detonation apparatus. The synchronizing signal from this circuit provided a timing accuracy of better than 1 microsecond, as determined by the distance of the trigger wire above the slit aperture section of the explosive and the occurrence of the luminosity peak on the oscilloscope sweep trace.

Several methods of photocurrent amplification have been used during the preliminary investigations. Initial tests were made with the phototube output connected through a megohm resistor coupled directly to the vertical plates of the cathode-ray tube. In later tests a single-stage dc amplifier with a type 6BA6 tube was used to obtain additional amplification for the input signal to the cathode-ray tube. Tests with unsheathed explosive charges have shown the necessity for higher amplification due to the lower luminosity of these charges and for a greater signal in the output stage to compensate for decreased deflection sensitivity of the cathode-ray tube.

In a verbal communication from James Taylor, I.C.I., Great Britain, to Dr. Bernard Lewis, it was noted that pronounced luminosity has been observed to emanate from inert pellets placed between pellets of explosives. A series of measurements showing the luminosity effects of layers of nonexplosive substances between explosive pellets has been carried out by us. Luminosity records have been obtained for the radiation emitted during passage of the detonation wave through pellets of sodium chloride and also through pellets of flake graphite and flake aluminum placed between explosive pellets. These layers of nonexplosive substances were more than 1 cm in length, permitting adequate definition of the radiation emitted by the material and excluding radiation from the explosive. PETN, tetryl, and TNT were used as the explosive substance, permitting investigation of the luminosity phenomena for a range of detonation pressure and temperature. Luminosity data were also obtained for plastic explosive compositions C-3 and C-4.

TABLE 2

Relative Luminosity for Detonation of  
Unsheathed Explosive Charges

Radiation Source	Pellet Density, gm/cm <sup>3</sup>	Relative Luminosity
PETN, micron diameter	1.50	1.0
Composition C-3	Plastic	3.0
Composition C-4	Plastic	3.0
Granular NaCl between PETN pellets	1.90	6.0
Flake graphite between PETN pellets	1.90	0.65
Flake aluminum between PETN pellets	2.00	1.5

Some results of the luminosity tests are shown in Table 2. The values of the relative luminosity were derived by interpolation of the signal amplification required for constant deflection on the detonation peak on the cathode-ray tube.

Most of our tests have been made with the explosive charge surrounded by air. Most of the luminosity observed in such tests probably originates in the early portions of the blast wave generated by the charge. The experiments with nonexplosive pellets provide support for this conjecture. The estimation of detonation temperatures will require the elimination of such effects by changes of the geometry of the experimental procedure and of the nature of the atmosphere surrounding the charge.

Since the slit aperture has a finite width, the resultant wave form is the result of integrated radiation, and the recorded peaks are not actual peak values. Reduction of the slit aperture with the necessary additional amplification and broader frequency response of the phototube-amplifier network will reduce the error of experimental determination of the detonation temperatures.

#### DISCUSSION

DR. KIRKWOOD: What about the preliminary results on the temperatures?

DR. MASON: Our difficulty is that they are all plus and minus 500 or more degrees. I can give you some examples. With a radiation slit width of one millimeter, for PETN density of 1.64 — five charges — 5750 plus or minus 550 degrees. PETN 1.18 — four charges — 6000 plus or minus 650. Tetryl 1.6 density — three charges — 4900 plus or minus 900. TNT (density of 1.56) 5500 plus or minus 350. Most of these traces have appeared in our quarterly reports and these temperatures have been in there. There is one exception to this which at the moment we can't explain. With the use of inert pellets, we get reproducible temperatures in the inert material, plus or minus 100 degrees, but we haven't explored that far enough yet to say any more.

QUESTION: What approximately are the shock temperatures in the pellets?

DR. MASON: About the same as those for the explosives.

DR. BRUNAUER: Is this the maximum range from the average?

DR. MASON: You get four temperatures from each stick, and this plus-minus represents the deviation between the four temperatures you get by comparing the intensity ratio from the four phototubes.

DR. KARTROWITZ: Have you taken any spectrograms of the light?

DR. MASON: Not yet.

DR. KARTROWITZ: I was wondering if perhaps some of the trouble in reproducibility had to do with the fact that the radiation is not black body.

DR. LEWIS: We feel it is necessary to study the quality of the radiation.

DR. BRUNAUER: Do I understand right that the detonation temperature of TNT was around 5000 degrees or something like that?

DR. MASON: For the TNT we have two values. For density of 1.56, 5500 plus or minus 350; density 1.29, 4850 plus or minus 400. I might say, I think Dr. Lewis will agree, due to the question as to whether we have true black body radiation—we don't like to commit



ourselves to saying these are the temperatures. We are skeptical of the translation of these luminosities into temperatures at the moment.

DR. BRINKLEY: I think perhaps it should be emphasized a little further that we are not certain that we are actually seeing the detonation waves. Until the effects have been thoroughly investigated in a systematic way, that question can't be answered. This explains our reluctance to call these detonation temperatures. We suspect that they are actually blast wave temperatures. For this reason, we have to be a little bit careful of interpreting the results. Before committing ourselves, we'll have to complete a greater number of tests.

MR. STREAU: You mentioned that you did something with powdered aluminum. Did you ever use ordinary cast or raw aluminum and see what happened with that?

DR. MASON: I believe that is contemplated on the program. We just haven't got to it yet.

DR. LAWRENCE: I wonder if I might refer to the work carried out by the Hercules Laboratory in 1944 when J. G. Fox devised a spectrographic method of measuring detonation temperature. He carried out quite extensive tests in order to separate the detonation luminosity from the shock-wave luminosity and found, for example, that where he had an air surround he got very much more intense luminosity than when he had a propane or water surround. You'll run into trouble if you have air bubbles in the charge because you have the shock wave traveling through the air bubbles in the explosive column as well as outside. It was also our impression that probably most of the luminosity came from the light scattered by explosive particles in front of the detonation, and this can be very strikingly illustrated. For example, Fox found that the temperature he was getting for tetryl was very much out of line with those he was getting from other explosives. Then he found that if he measured the light-transmission curve for tetryl and then corrected for the color of the tetryl, it brought the temperatures in line with the other temperatures. Just as a check on this, Fox ran another experiment with Cyclonite which he dyed blue so that there was a change in the color of light which would be scattered, and this had the effect of raising the apparent temperature about 1000 degrees K.

To come back to the effect of shock waves, we didn't actually measure any temperatures on these, but, qualitatively, their temperatures were very much higher than the detonation temperatures. As I say, we did carry out a good deal of work to try to eliminate the shock-wave luminosity in the surround and we would, for example, evacuate a charge and replace the air by propane as far as we could and then if desirable carry out the detonation under water using great care to get rid of all air bubbles which might carry shock waves.

We also carried on work with nitroglycerin where we felt that luminosity could be nothing but detonation luminosity. You could, if you wished, dye it black to cut down the scattered light. As I recall, the temperatures that Fox got were not unreasonably different from the calculated temperatures of Kirkwood and Brinkley and some others. But the main point I want to make now is that it is of very great importance to eliminate the shock-wave luminosity.

Fox carried out a number of experiments which indicated that the radiation was black body, but we all felt that more conclusive evidence was needed on that point.

DR. WALKER: The questions I was going to ask have been very well covered by the last speaker. I was going to ask what arrangement was made to eliminate luminosity of shock wave. What was the nature of the surround in these explosive charges? And the other thing—I was merely going to remark that in our studies we have been using salt pellets, length of three quarters of an inch and diameter about the same as in the explosive charge, and viewing them in a propane atmosphere which substantially eliminates all the shock waves. The luminosity seems to occur simultaneously over the salt pellets and commences as soon as the detonation wave reaches the initial surface of the salt pellets.

DR. RINEHART: We have been studying the burning associated with the solid particles. In that connection you get aluminum oxide which is given off by the particle which burns rapidly. We have recently taken spectrograms and find that most of the spectrum is continuous but we have observed Al or AlO lines. While there is a background there is a lot of spectra associated with the luminosity and I think, when you take spectrograms, you may well find it to be the case.

DR. COOK: I wanted to make one remark in connection with what Dr. Brinkley said, and simply say that I wonder if we can ever measure a detonation temperature directly from luminosity. When we realize that the detonation temperature corresponds to the back of the reaction zone and that, because of opacity, this will be different from this observed luminosity and also from that observed from the side, due to an edge effect, it certainly will make one wonder if he is ever going to measure detonation conditions directly from any such means. Of course, the best we can do is to measure what we see. Perhaps a technique for evacuation would help a great deal instead of trying to work in media that don't have much shock light intensity.

DR. LAWRENCE: You have to have a different medium. Evacuation doesn't do it.

DR. MASON: We tried evacuating the shots but it complicated the picture by putting more peaks in it.

DR. JACOBS: I would like to make further comments on that of Dr. Cook — whether you can expect to measure the detonation temperature since the only one that is important is that corresponding to the Chapman-Jouget condition. Unless you can measure at the Chapman-Jouget point you'll have nothing to compare with the hydrodynamic theory. If you consider the charge with a shock wave coming off the side, generally the luminosity of the shock wave does not build up immediately so you do see a peak. I am quite certain that this is mainly the luminosity seen through the explosive charge ahead of the shock wave. Some of this luminosity comes by transmission through the unexploded charge. Some of it may be due to the air shock, but that is part of the detonation when we are speaking of a detonation wave, and therefore the air shock is just contributing to the head of the detonation wave and not a shock ahead of it and separate from it. It is one consecutive thing. The other point is this: In the bounding medium the pressure is very low because it is pressure of a shock in the outside atmosphere. Therefore, if you cannot see in very far, you're only seeing light due to reaction at relatively low pressure, and measuring temperatures of reaction at 500 atmospheres or thereabouts. The only way to get the pressure of a Chapman-Jouget condition is to increase the pressure at the boundary. One way of doing that is to fool the detonation by having the wave impact on a solid in such a way that no rear effect and no shock wave is transmitted back into the explosive charge. If you can devise a transparent boundary which will allow you to see a detonation in this way, it is quite likely that you can get something which resembles the Chapman condition. Under these circumstances you'll have a pretty good chance of measuring the detonation temperatures.



## RECENT WORK AT NOL

Donna Price  
Naval Ordnance Laboratory

## INTRODUCTION

This paper summarizes various research projects carried out at NOL during the last three years. For each project, an effort has been made to describe the reasons the work was undertaken, the results of the program to date, and the current interpretation or proposed use of the data. Of necessity, the description of each program has been condensed and simplified. Similarly much of the specialized equipment and facilities (bombproofs, cameras, probe-timers) developed for the experimental work cannot be included in this report. However, a bibliography of the original work is included and covers the complete programs.

## SENSITIVITY TESTS

The practical necessity of meaningful sensitivity tests is obvious. In an attempt to develop such a test, which can also be simply made, that will give test values on a continuous scale, and that utilizes a test charge of large enough dimensions to eliminate propagation difficulties, the wax-gap test (1) has been evolved.

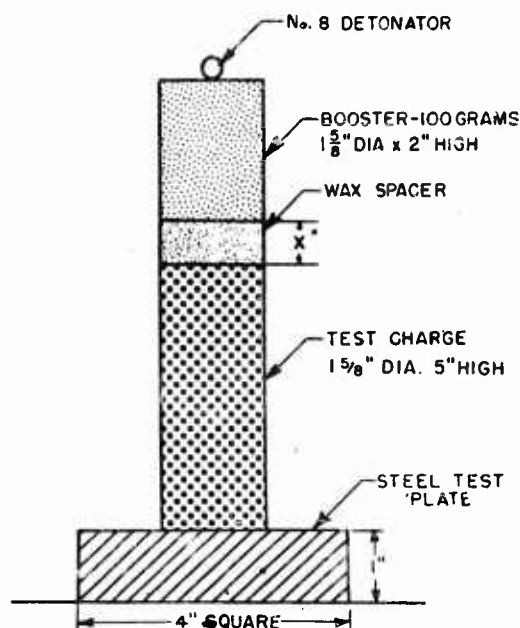


Figure 1 - Setup I for sensitivity test

## Wax-Gap Test

Figure 1 shows the standardized Setup I finally selected to satisfy these criteria. In each test, a 100-gram tetryl booster is used, and the acceptor charge is 1-5/8 inch in diameter x 5 inches long. The wax gap is varied by 0.05-inch increments during preliminary trials to locate the 50% point—the height of wax for which detonation occurs for half the trials made. Detonation is judged by the denting of the steel plate; failure, by lack of effect on the plate.

Table 1 shows typical results obtained with this standardized setup. You will note that the range of test values obtained was about a 2-inch height of wax. (Numbers in parentheses indicate the number of trials.) Statistical computations indicate a standard deviation of about 0.1 inch as a significant difference in test values.

The data in Table 1 have been arranged to show the usual sensitivity variation noted between cast and pressed charges. It should be pointed out, however, that in no case is the density of the cast and pressed material the same. Data obtained for various pressed densities of TNT indicate that extrapolation to the same density might eliminate

TABLE 1

## Wax-Gap Sensitivity Test

Explosive	Density (g/cc)	50% Gap (Inches)			
		Pressed		Cast	
Hydrazine Mononitrate	1.60	2.51	(14)	-	-
Pentolite	1.61	2.36	(13)	-	-
Pentolite	1.65	-	-	2.08	(12)
Fivonite	1.52	1.81	(13)	-	-
Fivonite	1.58	-	-	1.12	(12)
TNT	1.55	1.58	(12)	-	-
TNT	1.60	-	-	0.82	(23)
Composition B	1.69	-	-	1.40	(59)
Baratol 73/27	2.55	-	-	0.32	(12)

TABLE 2

## 50% Point vs. Spacer Material for Tetryl

Spacer Material	50% Gap (Inches)	
Acrowax B	1.89	(12)
Aluminum	1.90	(17)
Polystyrene	1.85	(15)
Copper	1.69	(13)
Oak	1.39	(17)
Air	5.04	(10)

this difference. Initiation difficulties on small quantities of highly compressed, highly confined materials in another study also point to rapid fall in sensitivity with approach to the crystal-density value.

Exploratory work on variations of the standardized setup has been sufficient to show that the test values obtained are definitely functions of the chosen conditions. Thus, while the results are fairly insensitive to the diameter of the acceptor charge, they are sensitive to booster height and, in this case, do not follow a linear scaling law. All results reported are, therefore, dependent on the standardized setup shown in Figure 1.

On the other hand, minor variations in the wax will probably have little effect. Table 2 shows typical results for wax and other materials used in the gap while testing tetryl. These data are interesting in several ways: the practically identical effect of wax and Al— and as Dr. Ablard's paper shows, we have considerable information about shocks in Al— and the very great difference between any solid-filled gap and an air gap. In the case of the solids, the initiation must result entirely from shock; no detonation products from the donor can make contact with the test charge. For an air gap, however, both shock and hot detonation products as well as the very different properties of the medium may play a role in the initiation.

The standardized wax test showed the expected decrease in sensitivity with addition of wax to the usual explosives (except for TNT). In general, it showed no effect of a temperature drop of about 100° C on sensitivity. Grit (sand and carborundum) had no effect on the test values. Although the test was satisfactory for most materials, a few were beyond the scale it covered. Thus, melted TNT and frozen nitromethane (NM) could not be detonated at 1-5/8-inch diameter even with a zero gap, although Fivonite (l) gave 0.47 inch and Fivonite (c) gave 1.12 inches. Since both TNT (l) and NM (c) gave detonations and partial detonations when tested at 2-1/4 inches diameter, it is probable that the standardized conditions chosen do not always avoid a diameter effect.

#### Air-Gap Sensitivity

The second sensitivity test that will be briefly described resulted from an investigation of critical variables in fuze explosive train components (2). Those results obtained for an air gap which are analogous to the wax-gap test work are the only portion of this program we will consider.

In this case, both the donor (booster) and acceptor are highly confined. The test arrangement, sensitivity Setup II, is shown in Figure 2. At the left is a brass cylinder bored to contain the initiator and the donor; at the right is a copper cylinder (with lip for ease in mounting) bored to contain the acceptor. The two parts are mounted and aligned on a lathe; the initiator (dextrinated lead azide) is fired by a condenser discharge.

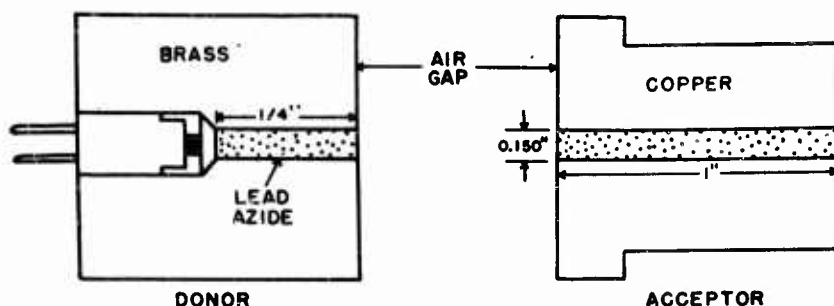


Figure 2 - Setup II for sensitivity test

The criterion for detonation is, in this case, spalling or change of diameter of acceptor container (end farther from donor).

Various factors affecting the test values have been studied. It was found, for instance, that the critical air gap increased with increasing donor diameter, increasing donor length, increasing donor density, and decreasing acceptor density.

Of these factors, the donor diameter and the acceptor density were found to have the greatest effects. Figure 3 shows three of these effects for shots on tetryl. Each curve shows critical air gap vs. tetryl density for the lead azide donor density indicated by the number written at the end of the curve. The acceptor diameter was kept constant at 0.200 inch. For the curves on the left, the donor diameter was 0.200 inch; on the right, 0.100. Doubling the donor diameter has increased the critical gap 4-5 times (top curves). In general, the gap length and donor diameter showed a linear dependence.

Figure 3 also shows the trend of increasing gap length with increasing donor density. A significant difference amounts to about 15% of the measured gap length. Hence, the crossing of curves shown at the left cannot be established without further work. The trend of increasing gap length with decreasing acceptor density is the conventional and expected one for tetryl; a similar trend was found in the wax-gap test for TNT. In the set of curves at the right, under these conditions, the sensitivity evidently decreases very rapidly as crystal density is approached.

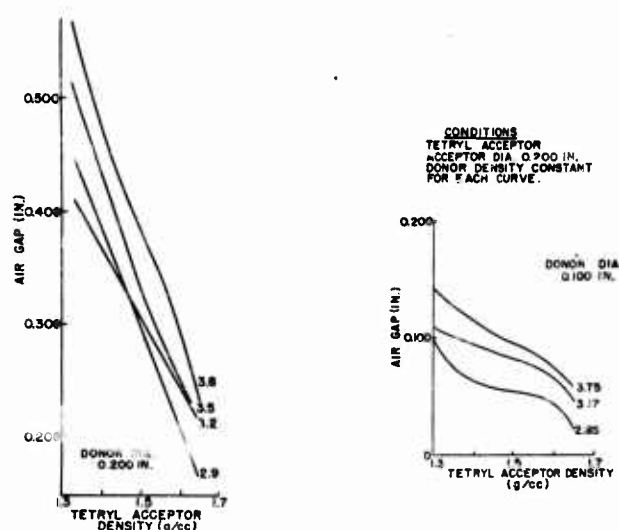


Figure 3 - Critical gap as function of acceptor density

After consideration of the factors affecting the test values, the standardized testing conditions finally chosen were:

Donor: Dextrinated Lead Azide

0.150-inch diameter x 0.500-inch length

Acceptor: 0.150-inch diameter x 1.00-inch length

Loading pressure: 10,000 psi

Number of shots for each determination: 15 - 20

Four materials have been tested in this manner as well as by the wax-gap test. The results and the corresponding densities are given in Table 3. In the three cases where both densities are known, they are nearly the same. This justifies some comparison of the air-gap and wax-gap lengths for these materials. This is shown in Figure 4 which does suggest a definite correlation between the two tests. The nature of the correlation will have to be determined by examination of more materials by both methods. The curvature of the line in Figure 4 indicates that initiation, in part by factors other than shock, plays a role in the air-gap test, particularly for explosives of lower sensitivity. Thus, if the trend RDX to tetryl were extended, no initiation of TNT would be obtained at zero air gap. Instead, initiation did occur and at an appreciable gap length. This agrees with the large difference for wax- and air-gap lengths found with tetryl in Setup I.

#### DETONATION VELOCITY MEASUREMENTS

In contrast to the sensitivity measurements, the detonation velocity of explosives can be determined quite accurately. Moreover, detonation velocity-loading density measurements provide data from which velocity of the medium surrounding the explosive (shell case, water, or air) can eventually be computed. Both their accuracy and their potential use to predict explosive effects make these measurements an important part of the present program.

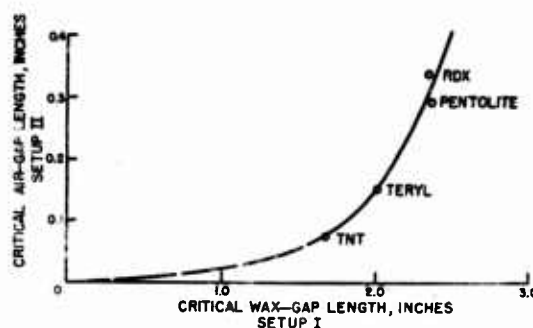


Figure 4 - Comparison of results of two sensitivity tests

TABLE 3

Critical Air Gap and Loading Densities of Pressed Charges

Explosive	Critical Air Gap (Inches)	Density Estimated for 10,000 psi	Density Obtained in Setup I
RDX	0.338	1.52	1.54
Pentolite	0.293	-	1.61
Tetryl	0.153	1.56	1.58
TNT	0.076	1.50	1.55

An immediate application of these data is a determination of the behavior of the detonation products. In Jones' recent formulation of the detonation theory (3), he expressed the detonation pressure as:

$$P = \frac{D^2 \rho_0}{\kappa + 1},$$

where  $\kappa$  is the adiabatic exponent, and

$$\kappa + 1 = (2 + \alpha) \left( 1 + \frac{\rho_0}{D} \frac{dD}{d\rho_0} \right),$$

where  $\alpha$  is a dimensionless quantity related to the variation with volume of the internal energy of the detonation products at constant pressure. The rate-density curve is linear at practical loading densities ( $\rho_0 \geq 0.8$ ). In this region, therefore,  $(dD)/(d\rho_0) = B$ , a constant, and we can define

$$\epsilon = \frac{\kappa + 1}{2 + \alpha} = \left( 1 + \frac{B\rho_0}{D} \right).$$

The quantity  $\epsilon$  is a measure of the departure of the detonation products from ideal gas behavior, and it can be determined entirely from the experimental  $D$  vs.  $\rho_0$  data. For an ideal gas,  $\epsilon = 1$ ; for explosives at a loading density of 1.6 and above,  $\epsilon$  varies from 1.5 to 2.0.

We have found that there is an empirical relationship (4) between  $\epsilon$ ,  $D$ , and  $Q$ , the detonation energy per gram of explosive. This may be expressed as

$$\frac{\left[0.04613 \frac{Q}{n} + 5.413 \times 10^{-5} \frac{D_c^3}{\epsilon_c^2} - 630\right]}{Q \rho_c} = 1.00 \pm 0.03,$$

where  $Q$  in cal/g and  $n$ , the number of moles of gas product per gram, have both been determined for an arbitrary decomposition with detonation products formed in the order:  $H_2O$ ,  $CO$ , and  $CO_2$ . The subscript "c" indicates that this equation is applied only at crystal density, but under this restriction twenty of the twenty-one sets of available  $D$  vs.  $\rho_0$  data fit it to within the indicated 3 percent.

The significance and extent of utility of this relationship have not yet been determined. It is being used at the present time as a test of consistency of the experimental data. For instance, an error in crystal density for one explosive resulted in a deviation of about 10 percent rather than 3 percent or less. Similarly, for another material, the  $D$  vs.  $\rho_0$  data used in the first trial did not lie on the curve for an infinite-diameter stick and the variation was again large. In both cases, accurate data reduced the variation to 3 percent or less.

The numerous  $D$  vs.  $\rho_0$  data now available for conventional explosives were used in evaluating  $\epsilon$  and in developing the empirical relationship just described. For the experimental program, however, attention has been directed to less familiar materials: hydrazine mononitrate (HN), nitromethane, and high-oxygen-content explosives.

#### Hydrazine Mononitrate

Hydrazine mononitrate is a solid (m.p.  $70^\circ$ ) and differentiated from the usual high explosives by (a) containing no carbon and (b) containing excess oxygen. Its detonation products would, therefore, be expected to contain a relatively high amount of water, and its study to yield information about the covolume of this gas as well as gas mixtures encountered in explosion products in general.

Rate-density studies were, therefore, carried out on HN with a conventional rotating-mirror camera setup (5). Although the material is quite sensitive to shock initiation, the charges were initiated by a two-inch length of tetryl booster. The results of this study, summarized in Figure 5, are both interesting and unexpected.  $D$  vs.  $\rho_0$  curves were obtained at four diameters. A conventional diameter effect appears at low  $\rho_0$  for diameters 0.5 inch to 1.0 inch. At high densities, however, there is also a diameter effect for diameters of 0.5 to 1.62 inches which results in maxima in these curves and actual failure of 2 charges of 0.5-inch diameter at a loading density of 1.63. The curves have been terminated at the highest density charge detonated except for the top curve which ends at crystal density. The only known similar diameter effect at high density is that reported for 60-40 Amatol for which the  $D$  vs.  $\rho_0$  curves are flattened at high density but show no maxima (6).

Coupled with the very large diameter effect, we would expect HN to show a curved detonation front. Preliminary examinations of the front profile confirm the presence of a marked curvature (as compared to the nearly planar front for Comp. B) and, for a 1-inch diameter, a greater curvature at

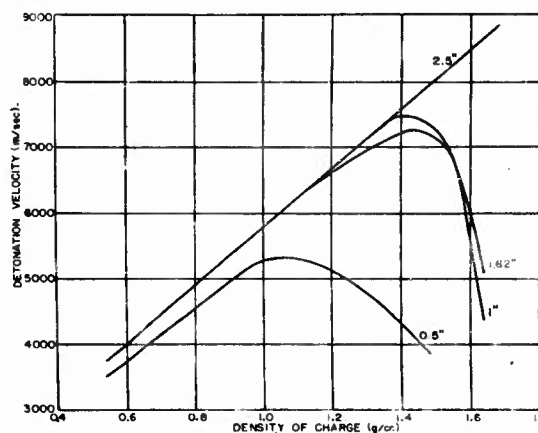


Figure 5 - Detonation velocity of hydrazine mononitrate

$\rho_0 = 1.6$  than at  $\rho_0 = 1.3$ . In addition, HN shows a high  $D$  and a steep  $D$  vs.  $\rho_0$  slope for  $\infty$  diameter. The high  $D$  is typical of oxygen-rich explosives, as the next project shows.

#### High-Oxygen-Content Explosives — Measurement of $D$ vs. $\rho_0$ on Small Quantities of Explosives

Most of the newer explosives of high oxygen content are available only in limited amounts. To obtain  $D$  vs.  $\rho_0$  data, therefore, it was necessary to devise a new method requiring but a few grams of the material for each test. It was found possible to obtain  $D$  measurements to about 5 percent by high confinement of small charges. Figure 6 shows the setup developed (7).

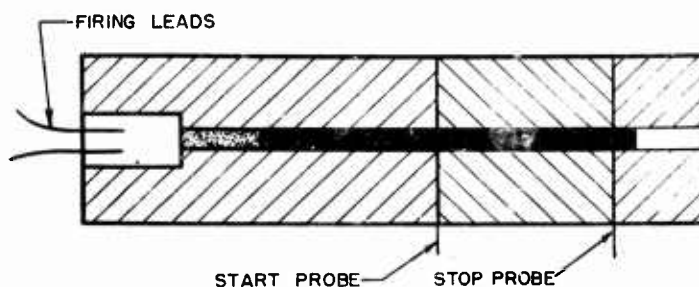


Figure 6 - Setup for measuring detonation rate on small charges

TABLE 4

$D$  vs.  $\rho_0$  Data for Oxygen Rich Explosives

Explosive	Slope ( $D$ vs. $\rho_0$ )	Crystal Density	$D_c$ (m/sec)
RDX	3533	1.82	8700 (9024)
Medina	4487	1.74	8772
BTNEU	5038	1.86	9091
BTNEN	3340	1.96	8719
TNEB	3459	1.78	8337
Nitromannite	3637	1.82	8441

crystal density,  $D_c$ . In fact, the actual rate values are probably higher than those shown under  $D_c$ , for the second value in parentheses for RDX is that obtained by extrapolating the optical data. It is about 300 m/sec higher than the value obtained by this probe method.

The extrapolated values of  $D$  at the crystal density are listed because they are only slightly higher than the highest rates measured. For example, RDX pressed to a  $\rho_0$  of 1.81 had  $D = 8650$  m/sec as compared to  $D_c$  of 8700; and TNEB pressed to  $\rho_0 = 1.77$  showed  $D = 8300$  ( $D_c = 8337$ ). On the basis of  $D$  values of high-density pressed charges, RDX, Medina, and BTNEN seem about comparable; BTNEU, slightly superior; and TNEB and nitromannite, slightly inferior.

Some work has been conducted on the detonation of nitromethane. It will not be described here. For those who are interested in this material, however, the factors investigated were diameter effect, sensitizers, and desensitizers.

It consists of three metal blocks with holes of 0.1-0.2-inch diameter bored through the center. The first block at the left contains the initiator followed by a half-inch length of lead azide, a half-inch length of PETN or RDX, and a half-inch of the test material. The middle block contains a one-inch length of the test material; and the end block, 0.2-0.3-inch of the test material. As indicated in the figure, the rate measurement is made by a probe timer on the one inch of material in the middle block.

This method is new, and there are many points yet to be investigated for specific materials. Complications can be introduced by such factors as different diameter, particle size, and sensitivity effects at different loading densities. The present tentative results are shown in Table 4. RDX is included as an example of more familiar materials. Medina and BTNEU show large values for the slope  $D$  vs.  $\rho_0$  as did HN, another oxygen-rich material. Although the slopes of the other three materials are more conventional, all five of these oxygen-rich explosives exhibit high detonation velocities at

## COMPUTATIONS

The present detonation theory permits calculation for a given material of  $D$ ,  $P$ ,  $\rho$ ,  $U$ , and  $T$  (detonation velocity, pressure, density, particle velocity, and temperature). The theory depends on the use of the usual detonation equations, the Chapman-Jouget condition, and an assumed equation of state for the gaseous detonation products. Such calculations, which are extremely tedious, have been carried out in great detail by Dr. Brinkley and others (8) for many of the more conventional explosives.

Dr. Snay and his associates have done a great deal of recent work on such computations (9). It is difficult to indicate in a short time how very helpful the resulting systematization and simplification has been. However, anyone who has made such calculations will appreciate the advantage of requiring only two computation sheets and the reading of three permanent diagrams to compute all parameters for a single explosive. These numbers are for an arbitrary decomposition and a known heat of formation for the explosive. One additional sheet is required for estimating the heat of formation (if unknown), one additional sheet for a mixed explosive, and several additional sheets for the calculation of equilibrium composition. The sheets are available in ozalid form and contain all necessary constants and operation directions.

TABLE 5 - TYPICAL COMPUTATION SHEET

DETONATION TEMPERATURE				
Iteration Process. Begin with an assumed $T_D$ , later enter result of previous step. Repeat until result does not change further.				
	First Step	Second Step	Third Step	Fourth Step
(a) $T_D = (T_D/T_B) \cdot T_B$				
(b) $n_g \bar{C}_v = A + B \cdot T_D$				
(c) $T_B = Q/n_g \bar{C}_v + 300^\circ \text{K}$				
(d) $T_B^{1/4}$				
(e) $x_B = h \rho_{eg}/T_B^{1/4}$				
(f) $1/\bar{C}_v = n_g/n_g \bar{C}_v \cdot 10^3$				
(g) $T_D/T_B$ Read from diagram.				
Final Result: $T_D =$ ; $T_D^{1/4} =$				
DETONATION VELOCITY				
(h) Read $I_D(x_B, 1/\bar{C}_v)$ from diagram.				
(i) $D^2 = \frac{n_g T_B I_D(x_B, 1/\bar{C}_v)}{M_0 (1 - \eta_{vs} \rho_0)^2} \cdot 10^5$	$D =$ m/s			
DETONATION PRESSURE				
(j) Read $I_2(x_D)$ from diagram using $x_B$ and $1/\bar{C}_v$ .				
(k) $P_D = \frac{P_g}{K} [T_D^{5/4} I_2(x_D)]$	$P_D =$ kbars			
Further Detonation Parameters:				
$U = P_D/\rho_0 D$ $D = \frac{\rho_0}{1 - U/D}$ $c_D = D - U$			Explosive	



Table 5 illustrates one computation sheet. In the iterative process, to find  $T_D$ , represented in the upper portion of the table, an initial value is assumed in (a). All other values are available from previous sheets, are computed by the operations indicated, or are read from one of the three diagrams. This sheet calls for three such readings, one from each diagram. The diagrams are:

- |                           |                     |
|---------------------------|---------------------|
| (a) $T_D/T_B$ vs. $x_B$   | various $\bar{C}_V$ |
| (b) $I_D$ vs. $\bar{x}_B$ | various $\bar{C}_V$ |
| (c) $I_2$ vs. $x_B$       | various $\bar{C}_V$ |

These are plotted on coordinate paper and a satisfactory device for rapid interpolation between curves has been developed.

For these calculations, the equation of state used was the Kistiakowsky-Wilson equation:

$$PVM = NRT (1 + X e^{\beta x}),$$

where

$$x = \frac{K}{T^{\alpha_{VM}}}, \text{ where } K \text{ is a covolume.}$$

The values  $x_B$  and  $\bar{C}_V$  are determined on the sheet shown in Table 5 and correspond to a constant-volume process (explosive  $\rightarrow$  products). From them the necessary functions for computing  $T$ ,  $D$ , and  $P$  are obtained from single readings of each diagram. The calculation is then completed by the simple operations indicated on the sheet.

In addition to this project, the same group is now in the process of finding the correct covolume factors from all available experimental data. Present results indicate that the correct values (especially for  $H_2O$ ) are different for those in current use.

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CONFIDENTIAL

## RECENT STUDIES AT THE NAVAL ORDNANCE TEST STATION

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### INTRODUCTION

The purpose of this talk is to describe work at the Naval Ordnance Test Station relating to the physics and chemistry of detonation. Generally, the level of effort in this field is not particularly high. The major efforts of the physicists and chemists at the Station are directed toward propellant rather than explosives investigations. The Station, in many cases, relies heavily on the work of other military establishments for the data which it needs in this work.

In the past, much excellent work was done by the Station's former Physics Division under the direction of Dr. W. M. Cady. Most of this work was stopped, however, when the group disbanded in September 1950. J. S. Stanton and his co-workers have published the results of several of their investigations as NavOrd reports and as technical memoranda of the Shaped Charge Working Panel. Cady, B. Cassen, Stanton, D. Marlow and others have published some of their results in the open literature.

Their more recent investigations include (1) an investigation of shock-wave reinforcement from simultaneously detonated charges (Stanton is continuing this work as an employee of Old and Barnes), (2) a study of the collapse of shaped-charge liners using luminous tracers, (3) the development of a number of micro-time techniques, particularly the development of a satisfactory Kerr cell camera, (4) a study of transient stresses in photoelastic substances, (5) an investigation of the decay of shock waves, and (6) the development of a method for the instantaneous measurements of velocity and temperature in high-speed air flow by the use of ultrasonic methods.

At the present time, three groups at the Station are actively engaged in studies relating to detonation. Dr. R. W. VanDolah's group in the Chemistry Division of the Research Department has been determining some of the physical properties of trinitrotoluene, and another group under Dr. A. L. Olsen in Chemistry has conducted a study leading to the design of a powder train delay. A group under my direction in the Physics Division of the Research Department has been interested in the reaction of metals to the impulsive type of loading generated by an explosive charge. In addition, we have been exploring various explosive systems with a view to obtaining very-high-velocity particles of predetermined size and shape. Mr. Guy Throner's group in the Rocket and Explosives Department is making valuable contributions in the way of novel, effective, and practical explosive systems and is conducting a number of shaped-charge studies on which he will report at this meeting.

In addition to these main efforts, miscellaneous studies are carried out at the Station from time to time by other groups or individuals. Recently, for example, Mr. K. S. Skaar, of the Rocket and Explosives Department, has undertaken the design of a waveguide that would give peripheral detonation of the explosive surrounding a shaped-charge cone. The design will be used to determine the efficacy of a waveguide in increasing the penetration of an antitank shaped-charge head for rockets. As part of the design study, the velocity of the pressure wave induced in maple by a detonating explosive charge has been determined experimentally. A value of approximately 12,800 ft/sec was obtained across the grain through 2 inches of maple. However, it appears that the velocity is an exponential function of distance through the wood since thinner sections give higher apparent velocities.

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Mr. Shook of the Physics Division is investigating the role that radiation plays in the ignition of propellants. It may turn out that some of the results will also be applicable to problems in the detonation of explosives.

## CHEMISTRY PROGRAM

### Physical Properties of Trinitrotoluene

Studies in the Chemistry Division at the Station under Dr. VanDolah brought on a discussion of the conflicting statements found in the literature as to the crystal system in which TNT crystallized. X-ray studies were made in an effort to resolve this conflict. It was found that TNT was polymorphic in character, showing both orthorhombic and monoclinic forms.

The observation of the polymorphic character of TNT tended to emphasize the meagerness of existing data on TNT and its reaction to varying environments. A program for a general study of the physical properties of TNT was then developed. This program was initiated with studies considered most likely to yield immediately useful information. The present work has been concerned with the phenomena of crystallographic transition and grain growth.

Experiments to determine the orthorhombic-monoclinic transition temperature led to the conclusion that this temperature is below 25°C. At lower temperatures, the rate of transition is so slow as to make the usual methods of detecting such changes inapplicable and work along these lines has been temporarily suspended. It has been found that various materials, including some which are normal impurities in TNT, are capable of affecting the rate of transition; some increase the rate of transition while others reduce the rate markedly, even at temperatures as high as 70°C.

TNT shows a marked tendency toward grain growth even at relatively low temperatures. Noticeable grain growth was observed in samples of TNT stored at room temperature for a period of two years. In general, it has been found that materials which, when added to TNT, increase the rate of transition from the orthorhombic to the monoclinic form also increase the rate of grain growth. On the other hand, those materials which reduce the transition rate reduce the rate of grain growth proportional to their effect on the rate of transition.

### Time Delay Element

The Chemistry Division has also developed, under Dr. Olsen's direction, a powder train delay element for use in the fuze of the 2.75 rocket. Several mixtures of explosive materials were tried. In the final design, the flash primer mixture within the delay cartridge is 53 percent potassium chlorate, 17 percent antimony sulfide, 25 percent lead thiocyanate and 5 percent TNT. The main body of the delay is 3,5-dinitrobenzoic acid, loaded incrementally under approximately 4000 lb/in.<sup>2</sup> pressure. The results of 36 test shots gave a mean delay of 491 microseconds, with a standard deviation of 164 microseconds. The delay cartridge is stainless steel, approximately one-half inch long with an internal diameter of 0.116 in. and a 0.042-in. wall. Its mechanical strength is such that it will not be ruptured by the explosion of the Mk 120 primer used to initiate the delay element. The results are purely empirical. No explanation of the mechanism responsible for the delay has been advanced.

## PHYSICS DIVISION PROGRAM

As mentioned above, my group in the Physics Division has been interested in the reaction of metals to the impulsive type of loading produced by an explosive charge. Our interests have been from both a phenomenological and a structural point of view. Much of

our work so far has been exploratory and qualitative. Generally, the action of an explosive is to produce fracturing and plastic flow. Our investigations are directed largely toward obtaining a better understanding of (1) the conditions that will lead to fracturing, (2) the type of fractures produced, (3) the conditions under which plastic flow will take place, (4) the structural character of the plastic flow that does occur, and (5) the mechanical properties of the materials that are significant under this type of loading.

One of the most instructive experiments has been that in which a small cylinder of charge is placed on the surface of a metal plate and then detonated. I will exercise a speaker's prerogative and discuss my own work in greater detail, devoting most of my time to a discussion of the results of these tests.

The experimental arrangement is shown in Figure 1. Composition C3, packed by hand into cardboard containers, has been used in all of the tests. Engineer's Special Caps were used to detonate the charges.

In such an explosive-metal system, three classes of fracture have been observed. These can be characterized in the following way: (1) scabbing, or spalling, the fracturing of the metal near one of its free surfaces which is relatively far removed from the area of application of the pressure (or stress) impulse, (2) fracturing of a type that probably can be attributed to sudden release of the applied pressure, and (3) fracturing that results from the lateral dilation that accompanies a vertical compression. Of these three types of fracture we have investigated scabbing most extensively and have obtained considerable quantitative data.

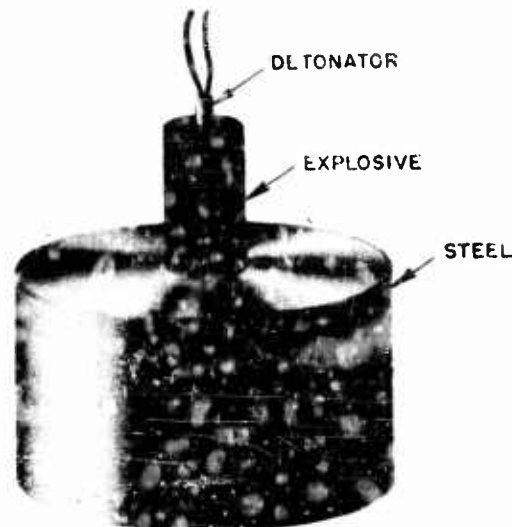


Figure 1 - Experiment arrangement

#### Scabbing

Geometrical considerations indicate that, if a plate scabs as the result of reflection of a high-intensity compressional stress wave at a free surface, the thickness of the scab ought to be governed principally by two factors: (1) the shape of the stress wave and (2) a critical normal fracture stress  $\sigma_c$  characteristic of the material acted upon. These same considerations indicate that the thickness of the scab ought to be equal to one half the distance within the wave that corresponds to a decrease in stress equal to  $\sigma_c$ . Scabbing should never occur when the maximum value of the stress within the wave is less than  $\sigma_c$ .

The method that has been employed to obtain stress information is somewhat similar to that used first by Hopkinson and later by others. Essentially, it consists of the experimental determinations of successive increments of momentums, i.e., areas under the stress-time curve, and, from these, construction of the complete curve.

Our experimental procedure includes determining the stress wave, measuring the scab thickness, and then deducing  $\sigma_c$  from these data. This has been done for plates of four thicknesses, 1-1/2 in., 2 in., 2-1/2 in., and 3 in. for each of five metals, annealed 4130 steel, annealed 1020 steel, 24 S-T4 aluminum alloy, annealed brass, and annealed copper.

A small cylinder (2 in. long, 1 in. in diameter) of explosive is placed on one face of a heavy metal plate and a small pellet (1/2 in. in diameter) of the same material on the opposite face, in line with the charge. A cutaway section of the arrangement is shown in Figure 2.

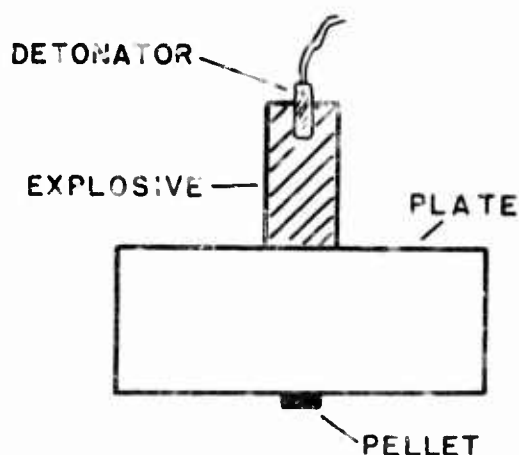


Figure 2 - Cutaway section

The plates were about 5 inches in diameter. Usually, five different thicknesses of pellets were used. Nominal thicknesses were 1/16 in., 1/3 in., 3/8 in., and 3/4 in.

The velocity-versus-distance curve obtained experimentally is then transformed to a stress-time curve through the relation,

$$\sigma = \rho cv,$$

where  $\sigma$  is the pressure (or stress) at a particular point,  $v$  is the particle velocity at that point,  $\rho$  is the density of the material, and  $c$  is the velocity of the wave.

A set of typical curves obtained in this way for annealed 1020 steel have been drawn in Figure 3. The curves have been plotted as histograms

since this type of plot represents most accurately the experimental data. The dotted smooth curve, sketched on each histogram, is the author's guess as to the probable shape of the true pressure curve. Similar curves have been obtained for annealed 4130 steel, annealed brass, 24S-T4 aluminum alloy, and annealed copper.

The deduced critical normal fracture stress for each of the five metals is listed in Table 1. No values for  $\sigma_c$  could, of course, be determined for those plates that did not scab.

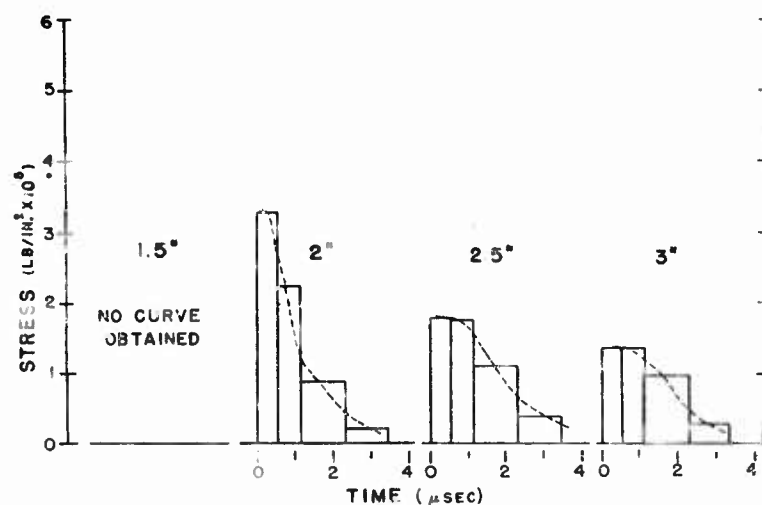


Figure 3 - Typical curves for 1020 steel

Two significantly different values of  $\sigma_c$  were found for 1020 steel. It is possible that the state of stress that exists in the region of the fracture at the time the fracture occurs may affect the value of  $\sigma_c$ . The higher value of  $\sigma_c$  was obtained for the higher ambient pressure. Such an increase in normal stress to fracture is in qualitative agreement with Bridgman's observations that resistance to cleavage fracture is increased by application of hydrostatic pressure.

TABLE 1

Comparison of Data with Clark and Wood's  
Critical Impact Velocities

Metal	Critical normal fracture stress $\sigma_c$ (lb/in. <sup>2</sup> )	Associated critical impact velocity (ft/sec)	Clark and Wood's critical impact velocity (ft/sec)
24S-T4	140,000	202	290*
Copper	430,000	277	235*
	400,000	258	
Brass	310,000	216	**
1020 Steel	230,000	120	100
	130,000	68	
4130 Steel	440,000	235	**

\*Calculated from static stress-strain curve.

\*\*Not determined.

#### The relationship

$$\sigma = \rho cv$$

can be used to associate a critical particle velocity with each critical normal fracture stress. Values of critical particle velocities obtained in this way are listed in the same table. These critical velocities and the critical impact velocities found by Clark and Wood agree rather closely. It seems not unlikely that, through further study, they can be shown to be physically equivalent.

#### Second Type of Fracture

Among the more unexpected fractures are those that can probably be attributed to the sudden release of the applied pressure. Numerous examples of this type of fracture have been observed. One of the most interesting is in the upper half of Figure 4, which is a photograph of a section cut from the middle of a plate that had been acted upon by a small cylindrical charge. A curved fracture resembling the arc of a circle was found about one-half inch below the surface of the crater. The shape of this fracture can be seen somewhat better in the lower half of Figure 4 which is a photograph of a section so cut that the material could be separated at the fracture.

It seems reasonable to assume that, in the present case, a very large portion of the energy of the compressional wave is stored in the form of a recoverable volumetric change. Upon release of the applied pressure, the body finds itself in an extremely unstable condition, with the energy very inhomogeneously distributed within it. The body will attempt to attain a stable condition as rapidly as possible. Fracturing, which is an energy-absorbing process, is undoubtedly one way in which the energy is dissipated.

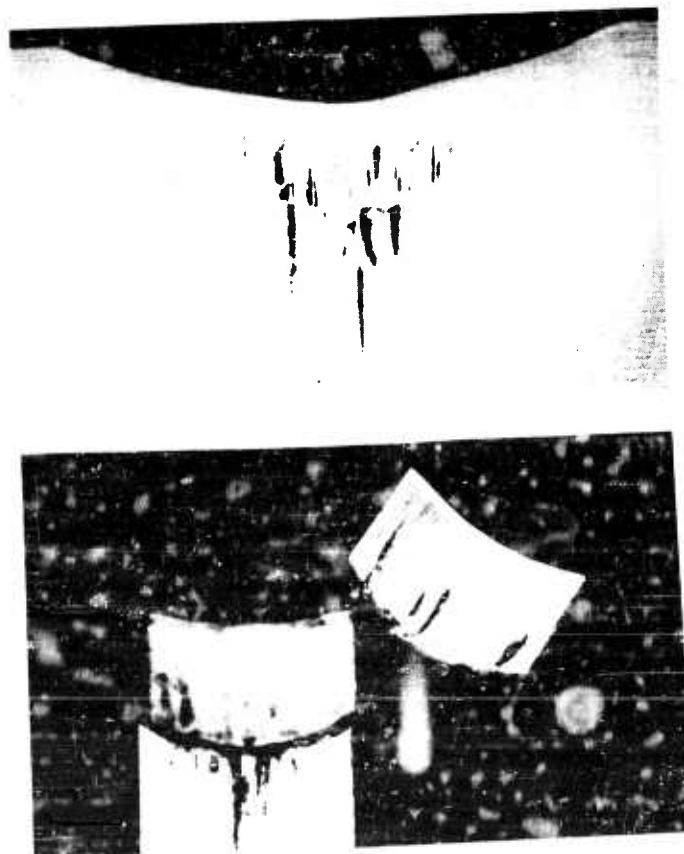


Figure 4 - Typical fracture due to sudden release of applied pressure

### Third Type of Fracturing

The over-all effect on steel of explosive cylinders of several diameters is illustrated in Figure 5. Many fractures, parallel to the direction of the applied stress, are seen to have been generated within the plate. The size, number, and extent of these are seen to increase with increasing charge diameter. They are brittle-type fractures. The tensile stresses that produced them probably originate in the dilation that accompanies application of a compressive stress. The tensile stresses will be expected to act normal to the direction of the applied compressive stress. Under slow loading rates, the material will flow plastically in a lateral direction. At very high loading rates, steel is known to behave more nearly as a brittle than a ductile material; hence, fracturing and not plastic flow can be expected to occur.

### Plastic Deformation

The plastic deformation that takes place within the plate is also being studied extensively, but time does not permit a detailed account of these studies here. Of particular interest, however, is a clearly defined region (Figure 5) that surrounds the crater and which, when etched heavily, contrasts markedly with the remainder of the section. The metal within this region has undoubtedly undergone severe coldworking. The hardness of the material within this region was found to be very much greater than the hardness of the material just outside of the region. The microstructures of the sections have also been studied. Typical structures are shown in Figure 6. The parent material is in the upper-left corner. The microstructure of the material below the highly worked region (bottom photograph in Figure 6) is characterized by the presence of a large number of Neumann bands or shock twins. Material within the highly worked region is shown in the upper-right corner. We do not yet understand the nature of the structural changes that have been wrought here. It appears probable that extreme shock-twinning occurs, but that, superimposed on it is extensive slip and some actual breakup of the grains into crystallites.



Figure 5 - Third type of fracture



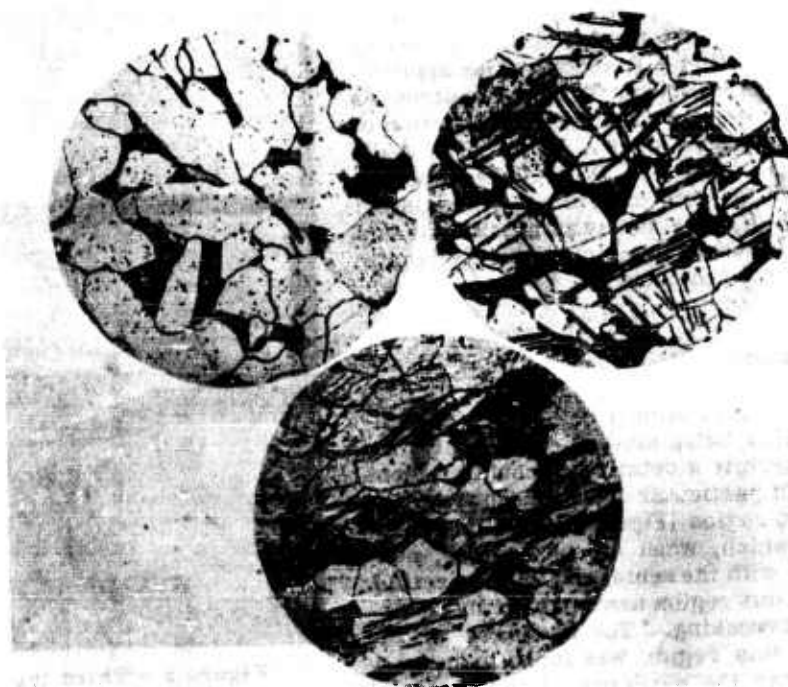


Figure 6 - Typical microstructures

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## SOME RECENT STUDIES IN CANADA

G. R. Walker

Canadian Armament Research and Development Establishment

This work was started back in 1943 when Dr. Hertzberg was at the University of Saskatchewan. He was asked by the National Research Council or one of its associated committees—that's the Canadian National Research Council—whether he would undertake some investigation into the field of explosives. He was advised by many people not to attempt it, because it was rather a difficult field and we had no facilities for setting off big charges, or anything of that sort, but he went ahead anyway.

I was with him as a student-assistant from the spring of 1944, and we carried on for about 18 months in that way. Dr. Hertzberg left the University in September 1945, and for the next few months we had some correspondence, but after that we were largely left to our own resources, and I carried on there until September of 1949. Since that time, I've been at the Canadian Armament Research and Development Establishment, or CARDE, as the initials would be pronounced; but up to the present time I haven't had any detonations there.

I have done some work, though, in attempting to study further the photographs which had already been taken at the University of Saskatchewan.

Now, very briefly, our facilities there were rather crude. We didn't have any concrete at all; we had very little steel in our bomb cell. It was mostly a wooden structure with earth banked around it and a few sand bags here and there. Our camera was one which Dr. Hertzberg had designed. We thought it would be rather suitable for this sort of investigation. It's of the rotating-mirror type streak camera. Its "F" number is relatively poor. It's about F-20. But its image speed is relatively large—5000 meters, which is comparable to detonations.

This camera, then, gives very good resolving power in the time direction—slits about a tenth of a millimeter wide, image speed, 5000 meters—that's 5 millimeters per microsecond. Also, it enabled us to study very small portions of the charge. We were setting off very small charges. And so the nature of initiation came to be one of the topics that we dealt with. I'll say something more about that in a moment.

We have done some work measuring detonation velocity. We were limited, of course, in the diameters of charges we could use, but ordinarily our accuracies were something like 0.2 or 0.3 percent. Rather considerably better accuracies were obtained for one liquid where we were using the differential method—measuring the velocity in this liquid at various diameters by measuring the difference between its velocity in various tubes and that in a standard tube filled with the same liquid.

The results there would probably be of some interest to a theoretician. We haven't done anything except to report them. Our work throughout has been almost entirely experimental reporting these things as we found them.

The question of detonation time—I am glad to notice that it has been given considerable attention here. We have photographed the zone of detonation following Dr. Lawrence's suggestion of using the stationary image, that is, using the image velocity as given by the mirror to compensate for the velocity of the detonation so that the image on the film is a

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stationary one. You make the slit wide or narrow, and the image remains unchanged—it merely becomes brighter.

And by measurement of such images, while I have no figures with me, and while such measurements are only now being made with a good microdensitometer, I can say roughly that the peak luminosity lasts something of the order of 0.02 microsecond and the total luminosity of detonation something like ten times that amount—perhaps not more than 0.2 microsecond.

Another thing which occupied some of our attention was observing Mach waves in detonations. And we observed all the usual things—that is, the incident wave, the reflected wave, and the Mach wave. These detonation waves are actually curved, as someone pointed out this morning. It is also true for shock waves. And the reflected detonation waves are extremely weak. But they do show up in the photographs.

Initiation—I can't very well say much about that, but we did study various high-density explosives, and a certain amount, also, of low-density explosives, initiated by detonator. We also have initiated them by the impact from what's now called a shaped charge, an aluminum-alloy jet.

But perhaps the more interesting results were the ones which I presented at the tripartite conference last fall at CARDE. I indicated at that time the means by which we could measure the velocity of the low-order detonations obtained when you have a jet, initiating high-density powders. The initial stages of the detonation are a low-order detonation.

Now, I indicated at that time how we had succeeded in measuring the velocity of this low-order detonation, which, incidentally, proceeds only down along the axis of the charge. It's not in itself visible, but by slightly indirect means we measured that velocity. I have no figures with me. The velocities were something of the order of 2000 meters, although they were not constant for one of the explosions. It would seem to depend on how far the lower wave travels.

We also have attempted to produce these low-order detonations artificially, in a manner somewhat analogous to what has been presented here at this meeting, by using a plain detonation wave, passing it through an inert material, and then passing the shock wave thus transmitted through the specimen concerned. We have used high-density solids and also liquid explosives. That shock wave passing through the solid or liquid seems to cause initiation not everywhere over its surface but just at one point on its surface, and from that point on the ordinary detonation spreads. There is a little more definite information in our reports dealing with these low-order detonation waves, including one or two attempts which we have made to cause two low-order detonation waves to intersect. However, that is something which hasn't been followed up very much as yet.

Now, I think I've said a few words about all the various things that we have poked around at, and I'm sure the impression you got is that we haven't really done much of anything in any one field but that we have just dug around a little bit and then tried to find out a little here and there as best we could.

Reports 1, 2, and 3 of our work, are, I believe, available from the National Research Council of Canada at Ottawa. And the other two reports, which I have numbered 4 and 5, are available as a single report from the Defense Research Board, also of Ottawa. Several other reports are in the process of preparation, and they'll be issued from CARDE as time allows us to get them finished.

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SESSION II

11 January 1951

CHEMICAL ASPECTS OF DETONATION

Bernard Lewis  
Bureau of Mines  
Chairman

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## CHEMICAL ASPECTS OF DETONATION

Bernard Lewis  
Bureau of Mines

The chemical aspects of detonation present a difficult subject for discussion. What we appear to be concerned with are those detonation phenomena that are neither predicted nor understood by the classical hydrodynamic theory. It would seem that our interest should gravitate toward solid explosives rather than toward gases. But we know generally more about gases than solids, so that it is perhaps more profitable to talk about gases.

It is quite natural to be interested in gas-phase detonation, since we have become acquainted with the striking phenomena of discontinuous progress and spin of the detonation wave in tubes filled with detonable gas mixtures; and it is intriguing to speculate how these phenomena might be harmonized with the classical concepts of the detonation wave. Von Neumann has provided us with the concept of the structure of such a wave, according to which there is a family of Hugoniot curves, each referring to a plane behind the shock front and representing stages of chemical reaction. Somewhere behind the shock front is the Chapman-Jouguet plane, in front of which the released chemical enthalpy is available for reinforcement of the shock front. There is nothing in the theory to indicate in detail the chemical and physical processes that occur behind the shock front in the various planes of Von Neumann. It is here that there is some leeway for further speculation.

We may introduce two different concepts of chemical transformation, each type being well-illustrated from other experience. One is exemplified by a gas element passing through the shock front and being thrown into a state of chemical reaction. In this process, diffusion and heat conduction between the various wave layers play a negligible role. One feature about the reaction may be assumed a priori, namely, the reaction rate is self-accelerating because heat is released and the temperature rises. It is, therefore, admissible to some degree to apply the concept of induction period whereby the element, after some time and corresponding distance of travel, will suddenly release the bulk of its chemical enthalpy.

In contrast to this type of process is the transformation in a combustion wave. Let us suppose that the reaction rate behind the shock front is at first very slow, permitting the element to travel some distance virtually unchanged, and that subsequently the reaction goes to completion within a distance of travel of only a few mean free paths. In that case, a zone is formed within which the rates of transport of molecules and heat by diffusion and conduction become of high order. The zone remains stable if the molecules of reactants that enter the zone can undergo complete reaction within a few (10 to 100) collisions.

This is no extravagant assumption because here the entering molecules are exposed to a combination of high temperatures and high chain-carrier concentrations. The concentration of chain carriers (atoms and free radicals) is large, even in the completely burned gas behind the zone, so that collisions with chain carriers constitute a substantial fraction of the molecular collisions. It is thus possible to visualize that the shock-generating thermal expansion takes place in a zone in which heat conduction and diffusion are controlling processes; in other words, a combustion wave is established behind the shock front. In contrast to the shock front which tends to be plane and perpendicular to the tube axis, the combustion wave is not stable toward fluctuations of particle velocity over the tube cross section. It must tilt and wander; and, since it is coupled with the shock wave, being the seat of the shock-generating force and thereby

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producing the medium within which it propagates, numerous phase relations appear possible which lead to more or less regular periodic phenomena.

Interesting possibilities arise when we consider that both types of combustion mechanisms operate simultaneously. There is no recognizable stability condition that limits the growth of area of the combustion wave. The chief limiting process that is operative in ordinary combustion-wave propagation along a tube is viscous drag at the wall, which is negligible here. Consequently, as time passes the wave area increases; and, since the tube walls provide confinement, the combustion wave assumes a tilted position of increasing angle of tilt. Simultaneously, since the total mass rate of burning is proportional to the area of the wave, the pressure signal to the shock front grows continuously, so that the detonation velocity increases. However, as the combustion wave becomes more tilted, certain gas elements "explode" before they have reached the combustion-wave front. This means a sudden additional release of chemical enthalpy which gives rise to a retonation wave. At the same time, however, the release of enthalpy in the pocket that is bounded by the combustion wave and the tube wall brings about a decrease of the area of the combustion wave and therefore a decrease of mass burning rate and weakening of the pressure signal to the shock front. The detonation velocity thus decreases and picks up again as the combustion wave oscillates between tilted and plane positions. Such periodic increase and decrease of the detonation velocity, interspersed with retonation waves, may be seen in the photographs of Bone and Fraser.

This concept leads further to an explanation of the "spin," which experimentally is known to be an intensely luminous spot that moves in a spiral path at high frequency along the tube and is capable of making a spiral trace in a silver film or in dust on the tube wall and to break a glass tube into spiral fragments, all of which is evidence of a local blast of hot gas. Such blast must occur at the upstream tip of the tilted combustion wave. Since conditions here are subsonic and since the gas expands in the combustion wave, there must be a refraction of the stream tubes entering the wave because of the continuity principle. This means that, at the tip, stream tubes expand on the unburned side of the wave and contract on the burned side and thus, because of refraction, direct a concentrated blast of hot gas against the tube wall. There is no stability for such a spot on the tube circumference, and it is quite readily imagined that the spot travels around the circumference. Since the whole disturbance moves forward, a spiral track is traced.

Whatever the merits of these suggestions, they certainly point to an interplay of chemical and physical aspects of detonation. The fact that spinning detonations seem to be generally observed as one approaches either limit of detonability lends further support to this interplay of chemical and physical factors.

In this particular theory the combustion wave would coincide with the Chapman-Jouguet plane. It is quite imaginable that, in other cases, the reaction mechanism is such that the reaction is more or less quenched before the total enthalpy is released, so that reaction is not complete in the Chapman-Jouguet plane. Thus only part of the chemical enthalpy is available for the detonation process. This at any rate might explain the occurrence of several detonation rates that have been observed with nitroglycerin and other high explosives. Incomplete reaction in front of the Chapman-Jouguet plane must also be a determining factor in the limits of detonability. It presages considerable difficulty in the search for a satisfactory theory of the limits of detonability, but perhaps it is less formidable than a theory of the limits of inflammability. In this connection I venture the opinion that, as difficult as the problems of detonation processes may be in their finer details, they probably are always considerably simpler than the formidable problems associated with combustion-wave propagation.

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## NONSTATIONARY DETONATION WAVES IN GASES

George B. Kistiakowsky  
Harvard University

Since 1946, my interests, as far as personal research goes, have been limited to gases which in comparison with actual explosives are rather feeble phenomena. But I agree with Dr. Lewis that in some way the study of gases is very useful, even as far as the understanding of solid explosives is concerned.

I'm afraid, however, that the particular phenomena which I wanted to discuss this afternoon happen to be very closely related to the detonation spin which Dr. Lewis has just discussed and don't have a parallel in solid explosives, largely because the phenomena in gases are very strongly influenced by the walls—the confining walls of the tube in which the experiments are made.

Dr. Lewis has mentioned that it's well known that these fluctuating detonation waves, so to speak, and spinning detonations, occur near to detonation limits. In our effort to understand the effect, so to speak, of chemical variables—reaction rates and such—upon detonation phenomena, we came upon the very old work of Dixon, who claimed that in very dry carbon monoxide mixtures, detonations were very seriously slowed down. Later work seemed to discount it.

Now, from the study of flames in carbon monoxide-oxygen mixtures, it's been definitely established that water or hydrogen are needed as intermediates in the chain mechanism—presumably the hydroxyl radicals and the dry carbon monoxide-oxygen mixtures burn at much slower flame velocities and have high ignition temperatures. We have studied the detonations in such mixtures, and I might perhaps say a few words about how these measurements are taken.

Most of the velocity determinations were carried out in stainless steel pipes of 10 cm diameter. The flanged pipes were made in two sections, bolted together, with a thin cellophane diaphragm clamped between the flanges. The ends of the pipes were closed by flat plates bolted to the flanges. To insure airtight connections, grooves were machined in the flanges, into which fitted rubber "O" rings. All other connections were welded airtight. The first "initiator" section was 50 cm long and was normally filled with readily detonable acetylene-oxygen mixtures, which were ignited by a powerful spark in a spark plug screwed centrally into the end plate. The other, "experimental," section, 120 cm long, was filled with the gas mixture to be examined. The initial spark ignition of the acetylene-oxygen mixtures was found to change into a detonation within a distance of a few centimeters. The experimental mixtures were detonated, therefore, by a nearly plane detonation wave striking them through the rupturing cellophane diaphragm. This technique eliminated the long run-up distances required to produce detonation waves from flames in less readily detonable mixtures.

The sensitivity of the method is about two-tenths of a microsecond, and the gages are spaced 10 cm apart so that we can get successive velocity intervals to certainly one percent. Now, when we plot the velocity of the initial shock which triggers the gages against the distance along the pipe, we find that a moist, essentially stoichiometric carbon monoxide-oxygen mixture has a perfectly normal detonation wave to—within the accuracy of the measurement—a few tenths of a percent.

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However, careful drying changes the phenomena completely. It becomes very difficult to initiate the wave. The wave has to be very definitely overdriven by a rather strong initiating mixture to get anything self-propagating in the dry mixture. And secondly, the velocity undergoes essentially periodic pulsations, of which we have observed, altogether, three, on a distance of two meters. If you wish to call it wavelengths, it's about 50 centimeters.

Incidentally, that ratio of wavelengths of these pulsations to the tube diameter is not very far from what has been observed in early work, particularly by the English school, using optical methods for observation and using narrow tubes. They worked usually with half an inch or, at most, one-inch glass tubes, in which case their moving film traces, instead of being straight, appear as wriggly records.

And again with these records are associated the so-called detonation spins. The variations in detonation velocity are by no means small. The normal excess, if you can call it normal at all, is maybe thirty percent in excess of Chapman-Jouguet. After these observations were made I started remembering the earlier work done by other students of mine in which detonating mixtures—hydrogen-oxygen, and so on—were under initiated. In that case, one usually observes a decaying shock, well under the Chapman-Jouguet value, then a violent rise in velocity. Of course, the gages being several centimeters apart, we cannot assert that it is an instantaneous thing, but it always occurs in the interval between two gages. The velocity becomes higher than Chapman-Jouguet—maybe sometimes as much as twice—and then drops to the Chapman-Jouguet value.

Furthermore, the thing that puzzled us then—two years ago—was that this surge was delayed about 50 centimeters which we now observed in the periodic situation. So I now think that the two types of phenomena have very close connections, and they have to do with initiation of detonations. And I want to speak now about a possible mechanism which I am very glad to see actually is very closely connected with what Dr. Lewis has spoken to us about just now. As he pointed out already, there is inevitable existence of an induction period between the initial shock and the onset of chemical reactions.

I was going to call this the Von Neumann zone, but actually I must warn you that it shouldn't be called the Von Neumann zone. Not so long ago I was reading a Russian article by Zeldovich published in a journal in 1940, which contains all the elements of the Von Neumann theory in detail. So if there is to be a name attached, I'm afraid it has to be Zeldovich rather than Von Neumann.

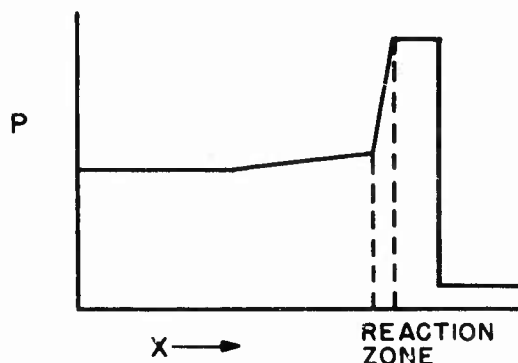


Figure 1

wavelength—again, only just a few tube diameters. So the whole phenomenon is tied into the tube diameter—that is, to the effects of the walls—and I imagine, although I'm not sure, that it has probably something to do with the turbulent boundary layer which must spread into the interior of the pipe. I don't know these details; they're certainly unclear to me.

So, as shown in Figure 1, we have to draw the pressure-distance curve because we have to remember that, if there is a zone where there is no chemical reaction and we are to have a stationary phenomenon, there can't be any pressure gradient. Now, what happens with these periodic detonations? I feel that it has to do with the fact that the reaction mixture is not sufficiently reactive to undergo chemical reaction within a very short interval after the compression by the shock front.

Now, what is "short"? "Short," I think, is a relative word in this case and relates completely to the tube diameter; these periodic fluctuations observed by others are only a few centimeters in



But now, let's see what will happen in either a poorly detonating mixture—that is, a mixture near the detonating limits—or in a mixture which has been deliberately made nonreactive—although an intrinsically good detonating mixture—by drying—like carbon-monoxide-oxygen. Let's start with the driven wave, which has been made to have higher intensity than the stationary Chapman-Jouguet intensity. At this point, we can assume that the initial shock is adequately strong, and in this case the temperature of the initial shock may be as high as 2000 or 2500 degrees Kelvin—enough to cause instant reaction. In other words, the profile is such that the chemical zone follows the shock almost immediately. But, the wave being overdriven, the energy dissipation by the wave is greater than the energy provided by the chemical reactions. And the wave must gradually decay to the Chapman-Jouguet stationary state.

When that happens, let's say at this point, the shock becomes not strong enough to insure the ignition of the gases in a region which is still nonturbulent and is free of wall effects. Consequently, the shock intensity begins to decrease, since it isn't being provided any more with a flame which follows it at the same velocity as the shock. The induction period must increase in duration, and the burning zone falls further and further behind the shock.

Well, if the flame front falls behind the shock and the pressure gradient becomes positive toward the shock, then the shock can be looked upon as a decaying shock, because there's a rarefaction wave behind it. And so, we have, then, the falling velocity of the shock; as the shock decreases in intensity, the induction period for ignition lengthens, and the flame falls further and further behind. The question now is, what causes sudden explosion? I feel the same way as Dr. Lewis does, although in detail a little differently, that it must be a fresh, spontaneous ignition. Because I cannot conceive of any mechanisms, even allowing for turbulence—after having had several discussions on the subject with several people—I can't think of any mechanism by which the same flame, after falling back into the turbulent region, could all of a sudden pick up such velocity that it could catch up with the shock.

And so I am inclined to think at present that a new ignition sets in. If the old flame is somewhere in the intermediate region, a new ignition sets in, so that there is a positive pressure gradient forward. Then presumably a new shock—maybe a pressure wave—then a flame front, which, of course, always means pressure drop inward; then a second flame which is moving backwards from this new spontaneous ignition region. I'll describe it as shock—to differentiate from flames—although there's no proof that they are real shocks since the whole thing is speculation.

And finally, somewhere farther back, is the initial flame. When the rear disturbance meets the original flame, that is the end of it. But when the forward disturbance reaches the original shock, you will have a very much higher mass velocity than is predicted by the Chapman-Jouguet theory, since you have a mass velocity due to the first shock and mass velocity due to the second disturbance—and, of course, once you're given the mass velocity you can see that the rest follows—that you have a sudden rise of velocity as detected by the gages.

If that is the correct interpretation of pulsating detonations, then the delayed initiations must be understood as follows. The first shock is moving forward, and is a decaying one. And by mechanisms which I freely admit I do not understand, our delayed ignition sets in, in a space between the initial shock and the air piston pushing the explosive gases forward, and this ignition spreads out. And when it catches the initial shock, we have, again, this extreme overshoot.

But now, since the explosive mixture is a good explosive, the phenomenon is a non periodic one, because the originally overdriven wave, insofar as its energy is dissipated, settles to a stationary regime of Chapman-Jouguet.

## DISCUSSION

DR. COOK: I'd like to call your attention to the fact that Parisot and Lafitte have observed the "spinning detonation" in amatol, and they found the same sort of thing that Dr. Kistiakowsky described here for gases.

DR. KISTIAKOWSKY: In a solid explosive?

DR. COOK: Yes, maybe Dr. Lawrence recalls.

DR. LAWRENCE: I'm a little bit hazy on that.

DR. COOK: Then I'd like to make one more comment. Becker, in his original treatment of this "spinning detonation," talks about dying out and reforming of the detonation wave—getting the effect that we get here, merely from the fact that we have a positive particle velocity in detonation, and a negative particle velocity in the deflagration process, so that you get a sort of a "retonation wave" effect. In other words, when the particle vector turns around, you have to conserve momentum by creating a "retonation wave" which now moves into the unreacted explosive and may be the reason detonation starts up again. I just wanted to add that as a possibility—and I believe it is a good possibility—that you have a periodic reinitiation of detonation in the "spinning detonation." But I think there's also a possibility that one obtains such a reinitiation by means of a "retonation wave" created as the particle vector turns around. In other words, perhaps this "retonation wave" is an explanation for the reinitiation suggested by Dr. Kistiakowsky.

DR. KISTIAKOWSKY: But how would that fit with the subinitiated good explosives?

DR. COOK: Well, this would always occur with materials that are right on the threshold of detonation. The threshold of detonation might be associated either with controlled boosting or controlled sensitivity. I think that may be the correlation, at least—that you require the detonation really dying out—but if you get right on that threshold of detonation by controlled initiation as well as by controlled sensitivity then you have a possibility of reforming an impulse of the detonation wave by means of a "retonation" wave.

In other words, we always talk of particle velocities being positive in detonation and negative in deflagration. The transfer from positive to negative would thus require a sort of "retonation wave." I'm just mentioning this as the Becker statement of it.

DR. KISTIAKOWSKY: May I say something? Just ten minutes before I started talking, Dr. Roberts gave me a report by Mooradian and Gordon which tickled me no end—because this is a measurement of the pressure phenomena—time-pressure curves in gaseous explosions. On inadequate initiations, when he plots the pressure-time curve, he finds a shock, and then another pressure pulse. The pressure pulse catches up with the initial shock—which to me is extremely encouraging, as suggesting that basically this idea of ignition far behind the front catching up with the front is not unreasonable.

DR. LEWIS: Why does it have to catch up with the front?

DR. KISTIAKOWSKY: Because, you see, this ignition is taking place in a medium with respect to which the shock velocity is subsonic. And therefore such a wave, which moves with at least sonic velocity, must catch up with it.

DR. VON ELBE: With respect to those criss-cross patterns in Bone and Fraser's photographs of spinning detonations, it seems to me that we must consider a mechanism of accelerating and decelerating wave propagation much as you have proposed, except that the frequency is higher than in the example described by you and that the zone of periodic change of reaction rate is at a much smaller distance from the shock front, perhaps a few centimeters

only. The spinning detonation seems to be a transition stage between ordinary steady-state detonation and the extreme case of discontinuous ignition that you have discussed. If I remember correctly, Bone and Fraser found spinning detonation in stoichiometric mixtures of carbon monoxide and oxygen to which a very small percentage of water vapor was added as a catalyst. When the water-vapor content was increased above some limit, the spin was eliminated.

Spinning detonation seems always to be observed when a mixture is weakened. It has been stated by Lafitte and his co-workers that spinning detonation or discontinuous progress is observed in any mixture when one approaches the limits of detonability.

DR. KISTIAKOWSKY: I might say that we have found, if not full periodicity, at least an irregular propagation of detonations in all such mixtures.

DR. HORNIG: Von Neumann points out that, with exponential reaction rates, it doesn't much matter whether you call it an initiation or let the reaction start immediately. But the rate of reaction after a finite period increases because of the exponential reaction rate with temperature behind the front as the reaction proceeds and heats up the gases, first slowly, and then very rapidly. And I think that's basically a correct point.

I'd like to comment, if I may, that I think that the Russians also have some data that bears on this question of the wall effect. I think it was also Zeldovich and the people associated with him who observed that, in cases of underinitiated gas mixtures where they got auto ignition after a period of travel, the auto ignition was very much speeded up by having rough-walled tubes as opposed to smooth-walled tubes. I think that ties in very well with what Professor Kistiakowsky has said.

DR. LEWIS: I have been looking over some old photographs that I took in 1930 in which an induction period is shown very clearly. These were Schlieren photographs. I wish to emphasize that one should not study detonation only with piezoelectric or other gages, because important phenomena may be missed. These photographs of gaseous detonation show clearly a separation in time between the shock front and a wave in which heat is involved, i.e., the combustion wave, of about  $10^{-5}$  second. This time represents a distance separation of the order of a centimeter.

DR. KAUFMAN: I just want to ask this question: We've heard a lot about the combustion wave. Now, I'm not sure - are there really two mechanisms possible for the propagation of this wave? In the Zeldovich-Von Neumann picture you have first a shock wave, which heats the gas to a temperature at which the induction period for reaction is very short - perhaps long enough to give the effect known as spin—but at any rate, very short. The energy released by the reacting gases then supports the shock wave. Now, is it possible to reach the same sort of picture with a combustion wave in the ordinary sense? I think you can prove quite simply that gradients which lead to diffusion and heat flow are totally insufficient to make the phenomenon stationary at the extremely high velocities that are actually observed. Therefore, diffusion and heat flow which present a sound mechanism for slow burning are not enough to explain detonation. I'm just wondering whether anyone has any suggestions. I can only see how a gas is heated rapidly to a temperature at which it's beginning to react, perhaps, with a short delay, but a combustion zone in the sense of the lower end of the Hugoniot curve I cannot picture at all because I don't see its mechanism.

DR. VON ELBE: I don't think that these gradients are necessarily small. It is conceivable that reaction goes to completion in a zone of only a few mean free paths width. In such a zone diffusion and heat conduction would not be negligible. The concept is that the gas passes through the shock front without undergoing rapid reaction until it reaches a zone within which reaction goes to completion in a few molecular collisions. In a sense, such a zone constitutes a combustion wave behind the shock front.

DR. KAUFMAN: Isn't the width, first of all, actually wider than the normal width of a flame—and also, isn't the shock temperature quite high?

DR. VON ELBE: No, the width of the zone that I have in mind is only of the order of a few mean free paths. The total width of the detonation wave is much larger and usually considerably in excess of the normal width of a flame. What is meant—and this is at present only an idea, not a proven fact—is this: A gas element crossing the shock front indeed attains a very high temperature and pressure, but this by itself is insufficient to initiate the rapid chemical reaction demanded by the detonation process. When neutral molecules such as CO and O<sub>2</sub> collide, nothing much happens even at those temperatures. The additional requirement is a high concentration of atoms and free radicals which act as chain carriers. When this concentration attains the order of the other molecular concentrations, the molecules of reactants, CO and O<sub>2</sub>, have a chance to react in a few molecular collisions. Let us now suppose that the reaction itself produces such high chain-carrier concentrations after an induction period which starts from the moment at which a gas element crosses the shock front. In that case the element travels for some distance in a state of high temperature and pressure with little chemical change; then suddenly the reaction goes to completion over a distance of only a few mean free paths. The resulting gradients of temperature and chain-carrier concentrations are such that heat conduction and chain-carrier diffusion are no longer negligible, and thus a zone of chemical reaction is formed whose propagation is controlled by heat conduction and diffusion in the manner of a combustion wave. This zone is unstable with respect to fluctuations of mass flow over the tube cross section; it must continuously tilt and wander. As it is coupled to the shock front by the fact that it furnishes the piston energy for maintaining the shock wave and is in turn dependent on the preheating and precompression occurring in the shock front, all manners of phase relations and periodic changes are imaginable. I think there is some evidence for such combustion waves in the photographs of Bone and Fraser.

DR. KISTIAKOWSKY: I agree with Dr. Von Elbe and also with Dr. Kaufman—that what is known as a spin is probably just a shock wave created behind the lagging combustion zone, the details of which I don't understand.

Now, more in detail to your remarks, Dr. Lewis. As little as I understand of hydrodynamics, I think the pressure gradient across the chemical reaction zone is defined completely by hydrodynamic considerations—the increase in volume, the heat released by the reaction, and the speed with which the reaction zone moves through the medium.

Now, as I see it, for instance, this pressure gradient across the chemical reaction zone, being caused by the considerations of propagation velocity and the nature of the reaction, is determined by the same laws in the reaction zone which exist behind the shock and what might be called the true deflagration of the flame.

There is one fundamental difference in my own thinking between the two phenomena. A normal deflagration is propagated by transport mechanisms—be it transport of chain carriers or transport of heat—but so long as you have these phenomena controlling propagation there is a certain finite velocity beyond which it can't accelerate—the limiting flame velocity. In distinction to that is a detonation, where the propagation of the reaction zone does not depend any more on the transport phenomena because a fresh reaction—if I may use such a crude expression—is generated in unreacted gas by an adiabatic compression of the shock so that the propagation mechanisms are different.

But I think the hydrodynamic laws of the zone itself are the same, whichever way it is propagated. But of course, actually, for instance, say in the Chapman-Jouguet detonation wave, the pressure drop across the reaction zone is very, very much higher than it is in a normal flame going through, say, hydrogen-oxygen. In a normal flame it moves with the velocity of maybe a hundred or two hundred centimeters, as against a thousand meters or so in this enforced propagation of a detonation. And that permits, incidentally, a perfectly logical definition of the instant initiation of detonation by deflagrations. And that is the one

where the mechanism of propagation of a deflagration changes from a transport mechanism to an adiabatic heating by the shock wave.

DR. LEWIS: In the deflagration wave or combustion wave that trails the shock wave the same mechanism obtains as in ordinary combustion waves except that the entire reaction occurs in a very much shorter time, because the gas leaving the shock wave is prepared.

DR. KISTIAKOWSKY: I don't know that it isn't essential. In our initiation experiments, when a shock enters an explosive mixture from air, the detonation picks up right at the boundary and just goes on, and there's no preparation by the chain carrier.

DR. KIRKWOOD: In connection with the relationship between the Hugoniot curve and burning, I'd like to ask if any one believes any more that burning has anything to do with this lower part of the Hugoniot curve. That is, you don't have shock.

The facts are pretty clear that you must have reignition in order to have this pulsating phenomena, and I wanted to ask both Dr. Kistiakowsky and Dr. Lewis whether you had any detailed ideas of how this reignition works.

DR. LEWIS: This is explained in my introductory remarks, and it concerns an induction period for ignition of the gases passing through the shock front coupled with the phenomenon of flame tilt.

DR. KISTIAKOWSKY: A chemically controlled induction period? Then it won't work, because it would depend on tube diameter.

DR. LEWIS: Yes, it does depend on the tube diameter. The frequency in the spin phenomenon is a function of tube diameter.

MR. STRESSAU: The question I had was, when you get this effect—whether it's rotating or pulsating or what-not—in the large tubes, is it obtained under the same conditions of pressure and mixture, and so forth, as it's obtained in the small tube? Or is the critical point that divides the detonable mixture different in the small tube from what it is in the large tube?

DR. LEWIS: I don't think we know, do we?

## DURATION OF THE REACTION IN A DETONATING EXPLOSIVE

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## INTRODUCTION

The Hydrodynamic Theory of Detonation, with its concept that the detonation is a shock followed by a deflagration, has attained to a rather satisfactory stage of development and acceptance. In this theory the idea of a finite reaction time is present, but hydrodynamics and thermodynamics alone shed no light on the kinetics of the reaction even to the limited extent of establishing its duration. To gain knowledge about the reactions duration, one must consider explosives finite in one or more dimensions. Only in this way does one obtain observable parameters concerning the duration. This is true both in experiment and in the accompanying theory. As a consequence we have the observation that detonation velocity is a function of charge diameter, and we have H. Jones' and H. Eyring's theories to explain this dependence in terms of a finite reaction zone. Similarly we have the experimental observations and theories of failure diameter and of over-and-under-boosting. More recently we have acquired further knowledge of the reaction zone in the experiments suggested by R. Goranson on the impact of detonations on metal plates. This is to be described by Dr. Ablard.

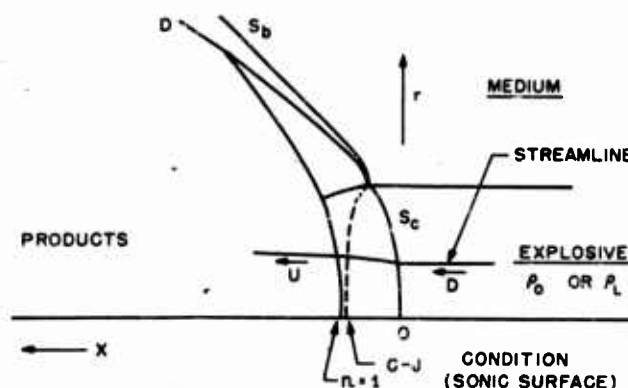
All of these experiments and theories relate an observable effect in bounded explosives to the presence of a finite reaction zone. In spite of the fact that a fairly large number of papers dealing with this subject have been written, there seems to be a feeling of dissatisfaction among the workers in the field concerning the significance and validity of the conclusions drawn in these papers. I think the trouble lies in our having insufficient understanding of the basic facts concerning the deviation from plane wave theory encountered when one begins to think of detonations which are finite in time and space. As a consequence the conclusions drawn by one author frequently contradict those of others and are even self-contradictory. As a result this phase of detonation theory is due for a thorough review. It is encouraging to find that some of the ideas of supersonic flow, shock waves, and rarefaction waves are catching on so that this mathematical field of hydrodynamics is being used, as it should, as a tool in the solution of problems of finite explosions.

## STEADY DETONATION IN TWO-DIMENSIONAL FLOW

I should like, now, to make a few remarks concerning the steady detonation conditions met in homogeneous explosives in two-dimensional flow, one dimension being the direction of propagation the other being thickness or radius. These can be termed slab and cylindrical symmetry respectively. I should like to consider the case when the explosive is bounded by an infinite low-density medium, for example air. These are the conditions under which most detonation velocity measurements are made. A profile of the detonation wave as I see it is shown in Figure 1. In this conception, which contains the combined and sifted ideas of many workers, the shock front is considered to be curved. The shock in the bounding medium is closely followed by the discontinuity between shocked medium and explosion products. The condition of reaction controlling the shock propagation in the explosive is drawn as the line designated as the general C-J surface. (The general C-J surface is here defined as that locus of points in the reaction back of the shock at which the outflow velocity is equal to the local sound speed.) The locus of complete reaction is drawn as the curve  $n = 1$ . As long as the shock front is curved, stream lines back of the shock can have radial component directly behind the shock. Several generalities can be stated concerning detonations in this configuration:



**Point 1. The front is generally non-planar.** At the boundary of the explosive, the pressure in the explosive shock is determined by the pressure in the surrounding medium. For air this pressure will be of the order of magnitude of 500 atmospheres, corresponding to an air shock velocity of about 7000 meters per second. Because of the low pressure, the explosive shock will be very weak and only slightly supersonic. The front will therefore be inclined at an angle whose  $\sin$  is  $C_0/D$ . The quantity  $C_0$  is the sound speed in the intact explosive. In this way curvature is accounted for at the edge. In general, curvature to the center will exist so long as the detonation is at a velocity below that for infinite media. The reason for this must exist if the detonation is slowed. The reason for this is the rarefaction to the rear. This is unlikely to occur at the sides. If the stream lines diverge at the front must be nonplanar. Near the characteristic distance of the distance to the sonic surface from the edge reaches the axis. I think it is probable that these distances will always be less than one. On the other hand, this is restricted only to being at or behind the sonic surface.



**Figure 1 - Detonation in a bounded explosive**

**Point 2. The reaction zone length is not constant throughout.** The very low pressure existing at the charge boundary with its accompanying weak shock in the explosive should mean that initiation temperature near the boundary is much lower than at the center. This, coupled with the lower pressure, should be reason enough to establish the existence of a lower reaction rate relative to the center and probably so low in many explosives that part of the explosive near the boundary may not react at all. In addition, the reaction temperature and products near the boundary will be quite different from those at the center. For this reason we might parenthetically remark it would appear undesirable to attempt to measure detonation temperatures in charges bounded by low-density media.

**Point 3. Curvature at the axis plus the extent of the completion of the reaction at the sonic surface determine the detonation velocity.** The outflow divergence of the stream lines will be determined by the curvature at the front plus the shock conditions met at the front. The latter are a function of the propagation velocity. Curvature alone cannot determine the propagation velocity since for weak shocks corresponding to low-rate detonations the curvature should be less than for some detonations propagating at higher velocity. This is due to the fact that the velocity can never be less than sound speed in the explosive. Low-rate detonations have less velocity difference between center and edge than do high-rate detonations. For very large radius of charge, most of the curvature will exist at the edge. As radius is reduced, curvature will move in to the axis and pass a maximum at the axis. If propagation continues, then reduction of radius will tend to reduced curvature as a result of the sonic limit.

## THE RELATION OF DETONATION VELOCITY TO CURVATURE AND REACTION

In conclusion I should like to present a simple analysis of the conditions near the axis of a charge. If we consider the front as having constant curvature near the axis, we can obtain simply the initial stream lines in this region. Referring to Figure 2., at the shock

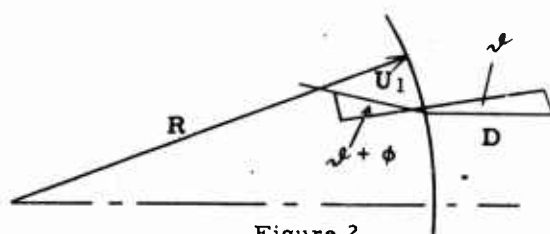


Figure 2

front we must conserve the tangential component of the flow through the jump so that

$$U_1 \sin(\nu + \phi) = D \sin \nu, \quad (1)$$

where  $\nu$  and  $\nu + \phi$  are the angles of the stream lines relative to the normal.

Therefore

$$\phi = \arcsin \left[ \frac{D}{U_1} \sin \nu \right] - \nu, \quad (2)$$

and for  $\nu$  small

$$\phi = \nu \left( \frac{D}{U_1} - 1 \right). \quad (3)$$

But  $\nu$  is given by  $\frac{r}{R}$  so that

$$\phi = \frac{r}{R} \left( \frac{D}{U_1} - 1 \right). \quad (4)$$

The radial component of flow velocity acquired in the shock is then given by

$$U_r = U_1 \sin \frac{r}{R} \left( \frac{D}{U_1} - 1 \right). \quad (5)$$

If we consider this radial component to be conserved in passing through the reaction zone, then

$$\Delta r(t) = U_1 \left[ \sin \frac{r}{R} \left( \frac{D}{U_1} - 1 \right) \right] t, \quad (6)$$

which for small angle  $\nu$  is

$$\Delta r(t) = \frac{r}{R} (D - U_1) t. \quad (7)$$

If we define

$$\alpha = \frac{A(t)}{A_1}, \quad A = \text{area} \quad (8)$$

we obtain for

$$\alpha = \left( \frac{r + \Delta r}{r} \right)^2$$

$$\alpha = \left[ 1 + \frac{D - U_1}{R} t \right]^2 \quad (9)$$

This therefore gives a pretty good approximation to the axial stream tube expansion in terms of the stream velocity immediately behind the shock and the radius of curvature of the shock front. We can now consider a perturbation on the plane detonation conditions in which mass and axial component of momentum are conserved in the stream expansion.



The initial equations become

$$D\rho_0 = \alpha U_x \rho_2 \quad (10)$$

$$\alpha p_2 - p_0 = D^2 \rho_0 - U_x^2 \alpha \rho_2 \quad (11)$$

$$E_2 - E_1 = \frac{p_2}{\rho_2} - \frac{p_0}{\rho_0} + \frac{1}{2}(D^2 - U^2) \quad (12)$$

These can be reduced to yield the following set of equations when  $U_x$  is assumed to equal  $U$ :

$$p_2 - \frac{p_0}{\alpha} = \frac{D(D - U)}{\alpha} \quad (13)$$

$$D^2 = (\alpha V_0)^2 \frac{p_2 - \frac{p_0}{\alpha}}{\alpha V_0 - V_2} \quad (14)$$

$$E_2 - E_1 = \frac{1}{2} \left( p_2 + \frac{p_0}{\alpha} \right) (\alpha V_0 - V_2) \quad (15)$$

where  $V = \frac{1}{\rho}$ .

These equations are identical to the equations for plane detonations, with the exception that the initial conditions are transformed to a new volume and pressure:

$$V_0^* = V_0 \alpha \text{ and } P_0^* = \frac{P_0}{\alpha}.$$

Since  $P_0$  is negligible for detonation of solid explosive, the value of  $\alpha$  can be determined from the detonation velocity provided we assume the C-J condition applies at the point of complete reaction. Under these circumstances the velocity of propagation of the finite charge is given by the velocity of an infinite charge of lower loading density. The equivalent loading density  $V_0^*$  immediately defines  $\alpha$ . The equations can be equally well applied to slab symmetry by redefining  $\alpha$  in terms of  $r$ . This changes the exponent of equation 9 to 1 instead of 2. The combination of detonation-velocity data with wave-front curvature therefore offers a method of determining the reaction time at the charge axis. If the sonic surface does not coincide with the end of reaction, more data will be needed to obtain the time to the sonic surface. For such cases, however, the propagation will be determined by the condition of tangency of the detonation velocity line (Equation 14) as defined above to a partial reaction Hugoniot which we can define in terms similar to that of Equation 15 as:

$$(E_2)_n - E_1 = \frac{1}{2} \left( p_2 + \frac{p_0}{\alpha_n} \right) (\alpha_n V_0 - V_2) \quad (16)$$

Of course  $\alpha_n$  is understood to apply to Equation 14 as well. In the limit of very low propagation velocity, such as in the low rate of detonation of nitroglycerine,  $\alpha_n$  will approach unity in agreement with statements which I made before. This viewpoint therefore includes an explanation, in principle, of the low rate of detonation frequently encountered for explosives.

It might be pointed out here that the Eyring point of view on the stream expansion considers the particles as moving radially toward the curved front. It is easy to demonstrate that this viewpoint imposes a slightly different assumption than mine on the stream lines. His point of view gives  $U_r$  decreasing with time as  $U_r$  is defined as proportional to  $(D-U)$  throughout the reaction zone.

When one inserts reasonable values for  $U(t)$ , the effect on  $(t)$  for the completed reaction turns out to be about 50 percent of that obtained if  $U_r$  is considered constant. The correct solution probably lies somewhere between the limits set by these alternate assumptions. It should be pointed out that both assumptions show the off-axis stream lines as curving toward lines parallel to the axis.

#### DISCUSSION

DR. ROTH: Do you know what  $(t)$  is, roughly I mean, from whatever data you have?

DR. JACOBS: We made one comparison on hydrazine mononitrate. We had already calculated the reaction time for particular charges of hydrazine mononitrate on the basis of the Eyring theory and on the basis of the Jones theory and have come up with answers of the order of magnitude of two microseconds. This results in similar answers. Namely, the theory is not different in magnitude but I think only makes it a little bit clearer as to what you're looking at.

DR. VON ELBE: This is somewhat off the point, but speaking about the curved detonation wave - when it comes to the end of the charge a jet is emitted, which I think shows up in numerous photographs, at the end of the explosive cartridge. And the front of the jet is extremely luminous. I just wondered—do you think that since the expansion occurs against a very low pressure so that there is little loss of internal energy, would it pay, perhaps, to make temperature measurements of this first gas, which shows up very well in photographs, and seek a relation between this temperature and the temperature in the detonation wave?

DR. JACOBS: Actually, this is aside from the subject, but I'd like to make a comment anyway. When you consider detonation reaching an end boundary, the detonation ceases as an entity and is replaced by a rarefaction wave running away from the boundary, a shock wave is sent out into the surrounding media, if there is a surrounding media, and a rarefaction wave eats into the explosion. Now if it takes a certain amount of time for the reaction to be completed, then a rarefaction wave begins to eat into it before the reaction has even started, and there is a great likelihood that one will have disturbed the reaction so much that it no longer will conform to the plane conditions that one associates with the detonation wave; and therefore any temperatures you measure will be associated with another phenomena and should not be interpreted as detonation phenomena.

DR. KIRKWOOD: To what order of  $1/R$  are these approximations on Hugoniot?

DR. JACOBS: The only approximations on Hugoniot, as far as I can see, is that, when you conserve momentum and you have a  $U_x$  and a vector  $U$ , I have substituted the vector  $U$  for the vector  $U_x$ . Therefore, if the angle is small, I think the order of difference is going to be quite small.

DR. STERNE: A question for my own information. I think you can adduce reasons—I don't know whether it's been done before or not—for part of the stability of a detonation. I'm thinking back to the previous talk of the oscillations. If for any reason the detonation process occurs at too high a rate, I think one can argue with considerable mathematical support that a rarefaction is going to eat away the detonation wave, reduce pressures, and get down to the Chapman-Hugoniot velocity. On the other hand, suppose the reaction velocity is less than Chapman-Hugoniot. What is the tendency that tends to bring the velocity up to the Chapman-Hugoniot? Can anyone answer that?

DR. KIRKWOOD: Dr. Brinkley and I have published a theory.

DR. STERNE: I think I remember the theory, but I don't know all the details of it.

DR. KIRKWOOD: It shows that if you've overdriven it, the pressure decreases towards the Chapman-Jouguet point. If you are underneath, you rise to it.

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## EXPERIMENTS ON THE TRANSITION FROM DEFLAGRATION TO DETONATION

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### INTRODUCTION

It is well known that, given the right conditions, many burning explosives will detonate. This transition from burning to detonation offers features of considerable practical and theoretical interest. In the handling and disposal of explosive materials, one often encounters fires. From safety considerations it is important to know when such fires can turn into explosions. On the other hand, one must know the conditions under which a properly initiated explosive will detonate, particularly in the case of primary explosives. Turning now to theoretical implications, it can be said that any complete description of detonation phenomena must account for these transitions. Conversely, the experimental study of the transition of burning to detonation provides a test for existing theories of the initiation of detonation. Our paper deals largely with this latter aspect.

Since we shall have occasion to refer to it, we would like to present here a brief outline of the theory of initiation championed by Prof. Kistiakowsky (1):

1. The flame from a burning explosive penetrates into the interior of the explosive mass, setting up pressure gradients due to the resistance to gas flow in the interstices between particles.
2. The rising pressure increases the deflagration rate. This further increases the rate of gas-volume production, which further increases the pressure, etc.
3. Gas flow from the center of deflagration, during the rising violence of burning, is accompanied by constantly rising backing pressure and temperature, which causes formation of shock waves.
4. Shock waves with their discontinuous rise in temperature and pressure greatly enhance the deflagration. The latter in turn reinforces the shocks. Eventually, all the grains of the explosive are consumed in one passage of the shock wave, and, therefore, the entire energy of the explosive can be utilized for the propagation of this wave. This is detonation.

When our work was begun, Dr. Cairns and Dr. Lawrence of Hercules Powder Company had already shown (2) that diazodinitrophenol, when initiated by a hot wire, shows a period of deflagration prior to detonation. Also, Prof. Bowden (3,4), of Cambridge, had published photographs showing that nitroglycerin and mercuric fulminate, initiated by impact, show similar predetonation deflagrations. We have extended these studies by obtaining rotating-drum photographs of primary and secondary explosives and smokeless powder under varying conditions of initiation, confinement, and packing density.

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## RESULTS AND DISCUSSION

## Primary Explosives

Mercuric Fulminate. - Of the primary explosives considered, mercuric fulminate best lends itself to our type of study. The transition from burning to detonation can be relatively slow, and the processes involved are sufficiently luminous to be recorded without difficulty. We found that confinement is the most important single factor in determining if the deflagration will change over into detonation. These trials were all made with the powder loosely packed in cellulose acetate tubes and initiated by a bridge wire imbedded in the explosive. Confinement was varied by altering the number of wraps of cellulose acetate used in making up the tubes.



Figure 1 - Mercuric fulminate—10-ply, 0.68-cm i.d. cellulose acetate foil tube; density 1.49 g/cc, aperture F:2.8

Figure 1 shows the transition for mercuric fulminate in a 10-ply cellulose acetate foil tube. The little luminous dot at "A" is probably due to the explosion of a few grains of powder adhering to the bridge wire, since the bridge wire fused by the discharge of a ten-cap machine will not register on film. Between "A" and "B" is a nonluminous zone lasting for about 300 microseconds. Deflagration starts at "B" and proceeds uniformly at a rather low rate up to point "C." Here an accelerating deflagration becomes visible. For the most part this deflagration moves at around 300 to 400 m/sec, but just prior to detonation at "D" it accelerates to 1100 m/sec. Detonation proceeds both upwards and downwards, indicating that it started in the interior of the powder column. There is appreciable afterburning. The total predetonation period lasts 8.5 milliseconds.

Figure 2 shows much the same sort of traces as Figure 1, except that all primary processes are much shorter, and the deflagration just prior to detonation proceeds at 1500 m/sec. The predetonation period lasts for 0.2 millisecc. If the confinement is increased even further, say with a glass tube, the preliminary processes can no longer be resolved.

Figure 3 is typical of the record obtained if there is just barely insufficient confinement to obtain detonation. Up to "C" the traces are similar to those in Figures 1 and 2. At "C" accelerating deflagrations of short duration are discernible. Considerable luminosity is observed at "D." Possibly this is caused by shock waves, formed by the deflagration, striking the stopper at the top of the tube. Again there is vigorous afterburning.

To summarize, we have observed nonsteady accelerating deflagrations and start of detonation in the interior. It is also possible that we have seen the manifestation of shock waves and their reflection in instances where no detonation regime existed.



Figure 2 - Mercuric fulminate—12 ply, 0.68-cm i.d. cellulose acetate foil tube; density 1.54 g/cc, aperture F: 2.5



Figure 3 - Mercuric fulminate--5 ply, 0.68-cm i.d. cellulose acetate foil tube; density 1.52 g/cc, aperture F:3.5



Figure 4 - Diazodinitrophenol—2 ply, 0.46-cm i.d. cellulose acetate tube; density 0.61 g/cc, aperture F:2.5

**Diazodinitrophenol.** - We were unable to obtain very significant photographs for the transition of burning to detonation for diazodinitrophenol because of the rapidity with which this takes place. However, Figure 4 shows an accelerating deflagration which rapidly turns into detonation.

**Lead Styphnate.** - Lead styphnate, the next explosive studied, is peculiar in that we were unable to obtain any detonation traces when initiation was by hot bridge wire. In every case, however, the container was completely destroyed. The luminosity was too weak to show up in an illustration. However, the luminous track occurs above the powder column. From the horizontal separation of the bridge wire and the start of the luminous track, we would judge that the process, whatever it is, occurs at an average rate of only several hundred meters per second. This would account for its lack of luminosity as well as for the well-known lack of brisance of lead styphnate. In agreement with this, Bowden(5) states that styphnate initiated by impact does not propagate at a rate greater than 700 m/sec. However, we found that styphnate, at a density of 1.56 g/cc, primed with 0.03 g of lead azide, detonated at 2,000 m/sec in a 5-mm i.d. glass tube. Also, when initiated with a spark from a condenser discharge (0.2 microfarad at 800 v), it detonated at 2,600 m/sec in a 7-mm i.d. tube at a density of 1.48 g/cc.

**Lead Azide.** - Even under the lightest confinement, lead azide detonates immediately upon contact with a hot bridge wire. We have been informed by Bowden(6) that, even with his new image-converter camera of great writing speed and tremendous light-gathering power, he has been unable to observe any deflagration prior to detonation for lead azide.

### Secondary Explosives

We know from previous experience(7) that PETN can be detonated by a high-energy condenser discharge. Since PETN does not burn readily unless it is subjected to moderately high pressures(8), we decided to investigate its transition from deflagration to detonation by obtaining photographs in glass tubes, with initiation provided by the discharge, through a bridge wire, of a 1-microfarad condenser.



Figure 5 - PETN—7-mm i.d. cellulose acetate foil tube; column height 13.5 cm, density 0.88 g/cc, initiated by condenser discharge 1 microfarad at 10,000 v

An example is shown in Figure 5. The deflagration is very short lived but quite apparent. The energy threshold for initiation is around 15 joules. At such high initiation energy, confinement and packing density (from 0.8 g/cc to 1.1 g/cc) have no apparent effect on the incidence of detonation.

Several trials with RDX failed to disclose anything but a deflagration trace with discharge energies of up to 50 joules. Bowden(5) was also unable to produce detonation in RDX and tetryl by condenser discharge.

### Smokeless Powder

With E. C. Blank Fire, an uncollicided single-base smokeless powder of great surface area, we obtained our most interesting photographs. Figure 6 shows a matchhead-initiated detonation. This photograph merits some detailed discussion. Deflagration is first visible at "A." Actually there is good evidence that nonluminous processes, if any, are of very short duration, since the vertical height AF corresponds very closely to the powder-column height. Deflagration proceeds at a rate of about 150 m/sec along the outer surface. Because of higher pressures, it is to be expected that in the interior deflagration moves up the powder column at a faster rate. This faster deflagration breaks out to the surface at "B." From Table 1 it is

apparent that the separation BC decreases with increasing packing density. This is in accord with expectations, since for compressed powders the deflagration encounters more resistance in penetrating into the interior and is forced back to the surface. BD shows a more rapid deflagration—of the order of 300 m/sec. Beyond D the deflagration begins to accelerate rapidly. At E it is moving at 900 m/sec, and just beyond E it goes over into a stable detonation. The trace EG indicates that the detonation started below the surface of the powder. For the sake of convenience, we have classified these processes as: initial burning (AB), accelerated deflagration (BD), low-order detonation (DE), and detonation (EF).

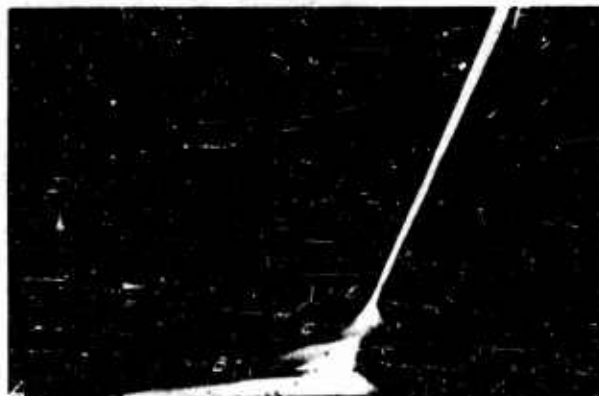


Figure 6 - Uncolloided single-base smokeless powder—8-mm i.d., 2.5-mm thick lucite tube; column height 53.2 cm, density 0.53 g/cc, matchhead initiated

TABLE 1  
Time Interval Between First Observable Burning and Breaking Out of Flame from the Interior of the Powder

Density (g/cc)	Initiated by	Time (Milliseconds)
0.82	M.H.*	0.05
0.66	M.H.*	0.12
0.56	M.H.*	0.17
0.53	M.H.*	0.18
0.52	M.H.*	0.35
0.51	M.H.*	0.37
0.54	M.H.*	0.57
0.52	C.D.**	0.57
0.46	C.D.**	0.62

\*Matchhead.

\*\*Bridge wire exploded by condenser discharge.

Detonations initiated by an exploding bridge wire from a condenser discharge are very similar in nature to matchhead initiations, as shown in Figure 7. The only apparent difference appears to lie in the initial deflagration rate, which is slower for the condenser discharge. This is not unexpected, since the greater area of the flame front and the higher initial pressure of the matchhead should outweigh the effect of the higher localized temperature of the exploded bridge wire. The energy threshold for detonation is about 10 joules.

The next two illustrations (Figures 8 and 9) show instances where deflagration fails to develop into detonation. The initial phases here are identical with those observed above.





Figure 7 - Uncolloided single-base smokeless powder—density 0.54 g/cc, column height 25.6 cm, initiated by condenser discharge of 1 microfarad at 5000 v



Figure 8 - Uncolloided single-base smokeless powder—density 0.55 g/cc, column height 25.5 cm, initiated by condenser discharge of 1 microfarad at 3800 v



Figure 9 - Uncolloided single-base smokeless powder—density 0.72 g/cc, column height 25.0 cm, initiated by matchhead



TABLE 2  
Rates of Various Processes in the Birth of Detonation in  
Single-Base Uncolloided Smokeless Powder\*

Density (g/cc)	Initiated by	Initial Burning (m/sec)	Accelerated Deflagration (m/sec)	Time of Accelerated Deflagration (M.S.)	Low-Order Detonation (m/sec)	Detonation (m/sec)	Delay from Initiation to Detonation (M.S.)
0.51	M.H.	100	360	0.05	880	2290	0.45
0.53	M.H.	140	ca. 300	0.08	920	2360	0.45
0.54	M.H.	136	350	0.08	-	Failed	-
0.56	M.H.	-	290	0.08	780	2430	0.31
0.56	M.H.	104	305	0.07	670	2380	0.44
0.66	M.H.	95	350	0.08	1010	2520	0.33
0.71	M.H.	100	320	0.06	880	2770	0.40
0.72	M.H.	100	270	0.05	810(?)	Failed	-
0.82	M.H.	-	340	0.08	670	2930	0.52
0.46	2800 v	78	214(?)	-	490	Failed	-
0.55	3800 v	47	300	0.07	330	Failed	-
0.42	4000 v	80	220	1.3	380	Failed	-
0.54	4400 v	52	370	0.07	660	2520	-
0.52	5000 v	23	360	0.08	810	2360	0.69
0.54	10000 v	60	-	0.08	700	2630	0.63

\*In 8-mm i.d. tubes.

Table 2 summarizes our observations. As already stated, initial burning rates are generally slower for "condenser-discharge" initiation. They appear to be independent of packing density. The "accelerated deflagration" rate, as well as its duration, appear to be independent of packing density and mode of initiation. For both of these regions, there is no distinction between failures and detonations. It is only in the next region, that of "low-order detonation," that any distinctions become apparent. If stable detonation is to result, the "low-order detonation" must apparently proceed at a rate of over 670 m/sec.

The difference in total delay from start to detonation between matchhead and "condenser-discharge" initiation is due to longer initial burning in the latter case. The observations of nonsteady accelerating deflagration, which goes over into detonation in a continuous manner, are, again, in excellent qualitative accord with Prof. Kistiakowsky's proposals.

#### Shock-Wave Initiation

We have made some exploratory experiments on the shock-wave initiation of E. C. Blank Fire. Shock waves of varying velocity were obtained from a piece of Primacord fuse placed at various heights above the powder surface. It is found that, in the packing-density range of 0.4 to 0.5 g/cc, the shock velocity threshold for initiation is around 4,200 m/sec under our experimental conditions. At higher densities no initiation could be obtained even with shock velocities of 6,200 m/sec. In Figure 10 we show a typical rotating-drum photograph. The



Figure 10 - Uncolloided shock-wave initiation of a single-base smokeless powder—40 cm. column of powder in 10-mm i.d. glass tube of 2-mm thickness, density 0.43 g/cc, spacing 2.5 cm

detonation track of the Primacord is observed at the top. The shock wave is a short, highly luminous portion, which may be detected immediately below the Primacord detonation. The horizontal trace of very high luminosity is due to the collision of the shock wave with the powder surface. To the left of this is a trace of much lesser brightness, which probably corresponds to the initial deflagration. Just below is a weak trace of an accelerating deflagration, and, finally, the detonation of the smokeless powder. This is, of course, quite analogous to the transitions shown previously.

The delay between the impact of the shock wave and inception of detonation of the smokeless powder varies with shock velocity.

Presumably, the high temperature of the powder surface, due to the impact of the shock wave, is a function of the shock velocity. If this temperature can be estimated and related to the observed delay, some very interesting conclusions about the kinetics of the processes leading up to detonation would be possible.

In conclusion, we would like to draw attention to the great similarity of transitions from deflagration to detonation under conditions of weak and strong initiation. Confinement appears to be the most important single factor which determines whether a deflagration shall die out or turn into detonation. In general, with the exception of lead azide and possibly lead styphnate,

our observations of accelerating deflagration which changes into detonation in a continuous manner are in excellent qualitative accord with the theory of initiation presented by Prof. Kistiakowsky.\*

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#### DISCUSSION

Mr. STRESAU: An interesting experiment was run two years ago. We didn't make any measurements of velocity, but we had some fairly highly confined columns of various explosives which we initiated, and then we observed the deformation. With mercury fulminate it was very easy to observe the transition of deflagration to detonation. The hole would stay the same size until you got to a certain point, and suddenly it got bigger. But with lead styphnate the hole had a very nice taper from one end to the other over a couple of inches.

Some day we intend to make some velocity measurements on this sort of thing. Incidentally, as the density was increased, this taper got more pronounced up to a point beyond which it reduced again, so that there was an optimum density for maximum expansion at the end. The standard column length was an inch and a half. It is probable that with long enough columns the increase in maximum expansion with initial explosive density would continue up to the highest densities.

DR. ROTH: How large a hole did you get with styphnate? Was it considerably smaller than azide or fulminate?

\*Note: After the presentation of this paper, Dr. Brunauer made the observation that some of the predetonation burning traces shown appeared to proceed at a steady rate and transition to detonation occurred in a stepwise fashion, which does not fit in with the theory of initiation because of the requirement of nonsteady deflagration and a continuous transition. While it is true that the initial portions of Figures 1, 7, and 8 appear to proceed at a fairly steady rate to within the resolving power of our camera, once the deflagration begins to accelerate, a detailed examination of the originals of these and other photographs shows that deflagration changes into detonation in a continuous manner. The curve traced by the transition is closely represented by a hyperbola with a steep slope on the detonation side and quite a flat slope on the deflagration side. The steady initial portions of these and other records could correspond to burning on the surface, i.e., to burning which has not penetrated into the interior of the powder and, therefore, can remain relatively steady because pressure could increase slowly and in a steady fashion.

Mr. STRESAU: Yes, it was always smaller.

DR. KISTIAKOWSKY: Didn't you also do some experiments with azide? Talk about them.

Mr. STRESAU: We are not sure of the mechanism involved. At high densities, about five or six percent voids or less, a reaction was observed quite different from normal detonation in its effect upon the container. The lower-density explosives, which are assumed to detonate normally, cause an apparent tension failure of the bore of the container. The failure is characterized by longitudinal cracks. Tension failure is to be expected, since the confining medium would be expected to have considerable outward momentum when the detonation wave has passed. The type of failure caused by the high-density explosives was quite different. It can best be described as looking as if it had been beaten out with a ball-peen hammer. As contrasted with the dull black coating left by the normal detonation, the bore had a high metallic luster. The color was white or grey, indicating deposits of lead and mercury on the brass container.

The propagation rate of the reaction which caused this latter type of deformation was measured and found to vary between 1,400 and 1,700 meters per second as compared with 5,000 meters per second for normal detonation of the same explosives loaded at a slightly lower density. It was suggested that the rate of propagations of this type of reaction might be related to the properties of the confining medium. No difference was detected between copper, steel, and aluminum in this respect.

This low-velocity reaction could be achieved only by using a fairly critically defined quantity of explosive. A slightly stronger initiator would cause detonation with a velocity in excess of 5,000 meters per second. A slightly weaker initiator would fail to initiate lead azide and would cause the mercury fulminate to burn at a rate of an inch per second or so. The burning was accompanied by the whistling of the escaping gases.

It was possible to detect the variations in loading density by the change in the note of the whistle. In fact, with care, one could count the increments.

In a similar set of experiments, it was found that, under some conditions, mercury fulminate could be made to burn for a certain distance and finally detonate. Audibly, this was characterized by a whistle followed by a report. In the sectioned containers, it was characterized by a length of hole in which no expansion was apparent, up to a point where a rather sudden change occurred, like a thistle tube. The length of the unexpanded section could be varied by changing either the loading density or the dimensions of the orifice through which the product gasses escaped.

These experiments may cast some light on the phenomenon known as "dead pressing." A "dead pressed" primary explosive has been defined as one which will not effect the transition from deflagration to detonation. Mercury fulminate is said in the literature to be "dead pressed" at 25,000 pounds per square inch loading pressure. In these experiments mercury fulminate, when confined only radially, burned for over a half inch of column length when pressed at only 5,000 pounds per square inch. When well confined in all three dimensions, it built up to detonation in less than 1/16 inch when pressed at 30,000 pounds per square inch. The growth of detonation is apparently a function of both loading density and confinement.

DR. LIDDIARD: How does the stepwise increase in velocity fit in with Dr. Kistiakowsky's ideas on the transition from deflagration to detonation?

DR. KISTIAKOWSKY: I don't see how to understand the step-wise acceleration of deflagration. Personally, I've always thought that the transition from a deflagration to detonation is—well, you might call it continuous or nearly continuous but substantially a sudden process. It may be the result of building a shock wave up to an intensity where it ignites the medium behind it. When it does, you will have sudden detonation. In other words, you will now have,

again, a change-over from a chemical propagation mechanism to a purely physical propagation mechanism caused by shock. But certainly, I do not understand how the deflagration rate should increase continuously.

DR. LEWIS: To what extent do you think that these step-wise changes are influenced by the positional coalescence of the compression wave or shock wave ignited from the containers, simply forming a node at that point and accentuating the building up of the energy at that point, and therefore causing the reaction to go to a larger extent?

DR. KISTIAKOWSKY: I think that's a very interesting point. That's very likely the case.

I might say we are not putting out results at a great rate, because all of this is done by just two graduate students, who are learning as they are progressing. They have been interested, in connection with the transition from deflagration to detonation, in acetylene-oxygen mixtures. And I want to draw a curve which really is a tickler.

If one draws a curve of the detonation velocity against composition of acetylene-oxygen mixtures, the curve rises quite steeply to a very sharp peak at almost exactly a one-to-one acetylene-oxygen mixture. The curve really looks like it has a discontinuity in it. This is an extraordinarily sharp maximum from which the velocity drops down very sharply and then levels off.

Now, we have not finished the Chapman-Jouguet calculations. So we do not know whether this is all consistent with the theory or not. For all I know, it may be. What we do know is this: that the Chapman-Jouguet calculation at this one point, somewhere around 80% acetylene, gives a velocity perhaps only 40 or 50 meters higher than observed under the assumption that the products of combustion are carbon monoxide, carbon, and hydrogen, allowing for hydrogen dissociation into atoms and equilibrium concentration of acetylene—for which there is enough thermodynamic data—not allowing for methane, ethane, and other such things but including solid carbon in the form of graphite. What you actually recover is a very finely divided carbon soot—and quite an active one, too.

So if you assume that this carbon has a few kilocalories higher energy content than graphite, which is certainly not an unreasonable number, then the Chapman-Jouguet calculation and the experiment will agree perfectly.

Now, this means that we have detonation where the chemical reaction zone must be controlled by diffusion processes, because these particles are of such magnitude that they must contain millions, if not billions, of atoms, which must somehow diffuse to these nuclei from the acetylene molecules.

We tried to see what happens if we assume gaseous carbon molecules, and that doesn't fit anywhere with the experiments. Quite recently we made some similar studies on cyanogen-oxygen mixtures, because Doeking and Schoen published a paper about a year ago which pointed out that in the mixture,  $C_2N_2 + O_2$ , according to their calculations, the only equilibrium components are carbon monoxide, nitrogen molecules, and nitrogen atoms. And consequently, the comparison of measured velocity with calculations should permit the calculation of the heat of dissociation of nitrogen, which has certainly escaped a lot of other methods for the last thirty years.

But we have made these first measurements, and we're quite worried that this objective may not be attainable because again we get a curve with a very sharp maximum—almost a singularity at this stoichiometric mixture for which the calculations are very simple, because there's very little else present in the thermodynamic mixture.

CONFIDENTIAL

SESSION III

12 January 1951

PHYSICAL ASPECTS OF DETONATION

John G. Kirkwood  
California Institute of Technology  
Chairman

CONFIDENTIAL

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## PHYSICAL ASPECTS OF DETONATION

John G. Kirkwood  
California Institute of Technology

I notice the title is "Physical Aspects of Detonation," although in looking over the titles it is very difficult to decide just how the distinction between chemical aspects and physical aspects is made. That is one of the things which makes the theory and experimentation in this field very interesting. We have a field in which it is necessary to call on the theoretical and experimental techniques of both chemistry and physics.

Since I'll have the opportunity to make some remarks this afternoon, I'll cut my introductory remarks short at this point and call on Dr. Stuart R. Brinkley of the Bureau of Mines for the first paper.

CONFIDENTIAL

## THE EQUATION OF STATE FOR DETONATION GASES

Stuart R. Brinkley, Jr.  
Bureau of Mines, Pittsburgh

## INTRODUCTION

The hydrodynamic-thermodynamic theory of the detonation velocity is based upon integral expressions for the continuity of mass, momentum, and energy across the detonation front formulated by Rankine and Hugoniot and upon the additional condition, stated independently by Chapman and Jouguet, that the stable detonation velocity is the minimum one compatible with the conservation equations. When supplemented by an equation of state and thermal data for the products of the detonation reaction, these relations are sufficient to permit calculation of the detonation velocity and particle velocity of the detonation wave and, in addition, the values of the state variables characteristic of the detonation gases immediately behind the detonation-wave front.<sup>1</sup>

Since the combination of high pressures, temperatures, and densities characteristic of the detonation gases from condensed explosives is inaccessible to independent experimental study, it is impossible to establish independently the correct form of the equation of state to be employed in the theoretical description of the detonation wave in such explosives. It has been customary to employ the theory in an inverted form, utilizing experimentally determined values of the detonation velocity to evaluate the parameters of an equation of state of assumed form. The equation of state obtained in this way can then be employed in the prediction of the detonation velocity of additional explosives, the success of such prediction usually being adduced as evidence for the correctness of the assumed form of the equation of state. For this program, several different forms, some of them unreasonable, have been employed, leading to different calculated values of the detonation temperature and pressure.

Knowledge of the equation of state for detonation gases is required in the more detailed theoretical study of the detonation wave and also for computation of the initial parameters of the shock wave generated in the exterior medium by the explosive charge. We shall survey briefly, and not at all exhaustively, several of the forms of equation of state that have been employed for the description of the detonation gases from condensed explosives, and we shall then consider the general problem of the evaluation of thermodynamic state from experimentally determined values of the detonation velocity.

## PROPOSED FORMS OF THE EQUATION OF STATE

Many investigations<sup>2</sup> of the properties of explosives have employed the Abel equation of state,

$$p(v - \alpha) = nRT/M, \quad (1)$$

<sup>1</sup>A summary of the hydrodynamic theory with references to the original papers has been given by G. B. Kistiakowsky and E. B. Wilson, Jr., OSRD Report 114 (1941) (Report PBL 32715, Off. Tech. Serv., U. S. Dept. Commerce). See also H. L. Dryden, F. D. Murnaghan, and H. Bateman, Bull. 84, Nat. Res. Council, p. 557, 1932.

<sup>2</sup>For example, Cranz, C. J., *Lehrbuch der Ballistik*, 2, 107 (1926); Crow, A. D., and Grimshaw, W. E., *Phil. Trans.*, A230, 39 (1930).



a simplified form of the van der Waals equation of state, where  $p$  is the pressure,  $v$  the specific volume,  $T$  the temperature,  $n$  the number of moles of gas contained in mass  $M$ , and where  $\alpha$  is an excluded volume, called the covolume. This form of equation has been widely employed in the interpretation of the pressure measured after the explosion of a charge in a closed bomb. The value of the covolume resulting from such studies is usually much less than that determined by considerations at ordinary conditions. When applied to the description of detonation gases,<sup>3</sup> covolumes determined arbitrarily so as to give the observed detonation velocity can have little physical significance. We note that this equation of state requires that the internal energy shall be a function of temperature only, the gas thus being thermodynamically ideal.

A more general form of the Abel equation of state has been assumed by Cook,<sup>4</sup> who employs the relation,

$$pv = nRT + p \alpha(v). \quad (2)$$

The function  $\alpha(v)$  is an arbitrary function of the specific volume, evaluated by comparison with experimental values of the detonation velocity. It was found that a single function was adequate for the representation of the detonation velocities of a number of different explosives, and excellent agreement between calculated and observed detonation velocities was obtained. As pointed out by Paterson, the arguments by which Cook justifies the assumption that  $\alpha = \alpha(v)$  instead of the more general assumption  $\alpha = \alpha(v, T)$  appear to be a posteriori in character. An equation of state of form essentially equivalent to Equation (2) has also been assumed by Caldirola.<sup>5</sup> Equation (2) implies that the gas is thermodynamically ideal, the internal energy being a function of temperature only.

Jones and Miller<sup>6</sup> have employed a virial expansion, terminating in the cubic term, explicit in the pressure,

$$pv = nRT (1 + bp + cp^2 + dp^3) \quad (3)$$

The virial coefficients, which they point out are properly functions of the temperature and gas composition, are assumed to be constants and are evaluated by a comparison between the theoretical relations and observed detonation velocities. The authors have published the results of calculations for the case of TNT only. Equation (3) implies that the internal energy is a function of temperature and pressure, varying as a power series in the pressure at constant temperature.

Kistiakowsky and Wilson<sup>7</sup> have employed a modified form of an equation originally constructed by Becker<sup>8</sup> on the basis of some theoretical considerations from high-pressure  $p$ - $v$ - $t$  data of Amagat on nitrogen. Becker's equation has been employed by Bridgman<sup>9</sup> for the representation of state data at 68°C on nitrogen to pressures of 15 kilobars. The equation is employed by Kistiakowsky and Wilson in the form

$$pv = nRT (1 + x e^{\beta x}), \quad (4)$$

$$x = k/T^{\alpha} v,$$

<sup>3</sup>As, for example, by Langweiler, V. H., Zeit. Techn. Physik, 19, 271 (1938)

<sup>4</sup>Cook, M. A., J. Chem. Phys., 15, 518 (1947). For discussion, see S. Paterson, *ibid.*, 16, 159 (1948), and Cook, M. A., *ibid.*, 16, 554 (1948)

<sup>5</sup>Caldirola, P., J. Chem. Phys., 14, 738 (1946). For discussion, see Brinkley, S. R., Jr., *ibid.*, 15, 113 (1947) and Paterson, S., *loc. cit.*

<sup>6</sup>Jones, H., and Miller, A. R., Proc. Roy. Soc. London, A194, 480 (1948).

<sup>7</sup>Kistiakowsky, G. B., and Wilson, E. B., Jr., *loc. cit.*

<sup>8</sup>Becker, R., Zeit. f. Physik, 4, 393 (1921)

<sup>9</sup>Bridgman, P. W., Proc. Am. Acad., 59, 173 (1924)

where  $\alpha$ ,  $\beta$ , and  $k$  are empirical constants. The parameters of Equation (4) have been evaluated<sup>10</sup> by comparison of the theoretical relations with experimental detonation velocities. Equation (4) was employed in calculating values of the state variables for a number of explosives and by Kirkwood and his collaborators in determining the initial parameters of the shock waves in water and air generated by the explosive charge. Equation (4) implies that the internal energy is a function of temperature and density, varying as an exponential in the density at constant temperature.

Calculation of the pressure, density, and particle velocity characteristic of the detonation velocity is relatively insensitive to the form of the equation of state employed for the description of the state of the detonation products, and the different equations of state predict values of the same order of magnitude. However, the calculated temperature is quite sensitive to the form of the equation of state employed. In particular, if the composition of the detonation products does not vary with the loading density of the explosive, the calculated temperature will increase with increasing loading density for equations of state such that the internal energy is independent of the density. If the energy of gas imperfection is significant compared to the internal energy of the ideal gas, the calculated temperature will decrease with loading density. These considerations are illustrated by Table 1, which lists a few calculated results for TNT.

TABLE 1  
Calculated Values of Detonation Temperature

Loading Density	Cook	Jones	Kistiakowsky and Wilson
1.0	3700	3800	3170
1.5		3400	
1.6	4170		3170

#### DETERMINATION OF THERMODYNAMIC STATE FROM DETONATION VELOCITY

It is instructive to consider the general problem of determination of the thermodynamic state of the detonation gases by means of the hydrodynamic theory using an experimental curve  $U = U(\rho_0)$ , where  $U$  is the detonation velocity and  $\rho_0$  the loading density. If the particle velocity is eliminated, the Hugoniot equations can be written in the form,

$$\begin{aligned} p - p_0 &= \rho_0 U^2 (1 - \rho_0/\rho), \\ H - H_0 &= U^2 \left[ 1 - (\rho_0/\rho)^2 \right], \end{aligned} \quad (5)$$

where  $p$ ,  $\rho$ , and  $H$  are the pressure, density, and specific enthalpy of the detonation gases at the wave front, respectively, and where  $p_0$  and  $H_0$ , the pressure and specific enthalpy of the intact explosive, respectively, are assumed to be given. The Chapman-Jouguet condition can be written in the form,

$$c = \frac{\rho_0}{\rho} U, \quad (6)$$

where  $c = [(\partial p / \partial \rho)_s]^{1/2}$  is the Euler velocity of sound in the detonation gases. Although it is customary to employ an equation of state for the pressure explicit in the temperature and density, this choice leads to considerable algebraic complication in the development of the theory.

<sup>10</sup>Brinkley, S. R., Jr., and Wilson, E. B., Jr., OSRD Report 905 (1942) (Report PBL 31088, Off. Tech. Serv., U. S. Dept. Commerce)

A more natural choice is to assume the existence of a relation,

$$\rho = \rho(p, S), \quad (7)$$

where  $S$  is the entropy. The usual program for determining an equation of state from experimental values of the detonation velocity is the formal equivalent of the determination of function (7), for given values of  $\rho_0$ ,  $p_0$ ,  $H_0$ , and  $U = U(\rho_0)$ , by means of Equations (5) and (6) and making use of the laws of thermodynamics. However, it is easy to show that the Jacobian

$$J \begin{pmatrix} \rho, H \\ p, S \end{pmatrix} \quad (8)$$

equals zero. This implies that the fundamental relations and given quantities are not sufficient for the unique determination of the thermodynamic state from detonation velocities alone but that the given relations can be satisfied together with any arbitrary function  $p = p(S)$ .

It follows that the degree to which an equation of state of assumed form reproduces the experimental data employed in the evaluation of its parameters cannot legitimately be employed as a criterion of the correctness of the form of the equation. Assumption of the form of the equation of state provides the additional assumption required to make the theory determinate. Selection of the form of the equation of state must be based upon additional evidence, either experimental or theoretical in nature, or, lacking such information, on considerations of plausibility.

#### ENERGY OF GASES AT VERY HIGH PRESSURE

Immediately behind the detonation front, the density is greater than that of the intact explosive, and the molecular volume is less than the excluded volume necessary for the application of an Abel type of equation of state. Under the high compression that exists in the detonation wave, it seems clear that the repulsive forces between the tightly packed molecules must make a significant contribution to the energy.<sup>11</sup>

In an important and informative paper, Jones<sup>12</sup> has employed considerations entirely independent of the detonation process to establish an equation of state of the detonation products, and he has employed the resulting equation in an *a priori* calculation of the detonation velocity for several loading densities of TNT and lead azide. In formulating an equation of state, the limiting case of very high pressure at a fixed temperature was considered, for which it was possible to construct theoretically the form of the equation of state in a way similar to that in which the equation of state of a solid is constructed. The parameters of this equation were fixed, for the particular case of nitrogen, employing Bridgman's data. A virial form of equation was employed to interpolate between high and low pressures. In this way the detonation velocity and values of the state variables were calculated without any reference to explosion experiments. The detonation velocities thus obtained were about 17% higher than the experimental values, but the variation of detonation velocity with loading density was very well reproduced. The calculated detonation temperature was a decreasing function of the loading density. These considerations lead to the prediction that the internal energy of the detonation gases is an exponentially increasing function of the density at very high pressures.

These considerations lend considerable support to the equation of state for detonation gases in the form proposed by Kistiakowsky and Wilson, Equation (4). This equation has the indicated exponential dependence of internal energy on density at high pressures, and at vanishing pressures it has the correct ideal gas form. It may be noted that, by adopting a suitable value of the

<sup>11</sup>H. Jones and A. B. Miller, *loc. cit.*

<sup>12</sup>H. Jones, Ministry of Home Security (Great Britain) Report RC-166 (1941)

parameter  $\beta$ , this equation can be made to agree closely with Hirschfelder's<sup>13</sup> virial form of the equation of state that has been successfully employed for the description of propellant gases.

### ESTIMATION OF THERMODYNAMIC STATE OF DETONATION GASES

Jones<sup>14</sup> has recently shown that

$$\left. \begin{aligned} p &= \frac{U^2 \rho_0}{(2 + \alpha)(1 + g)}, \\ \rho &= \rho_0 \frac{(2 + \alpha)(1 + g)}{(2 + \alpha)(1 + g) - 1}, \end{aligned} \right\} \quad (9)$$

where  $g = d \log U / d \log \rho_0$  can be determined from an experimental curve  $U = U(\rho_0)$  and where

$$\alpha(p, \rho) = \left( \frac{\gamma C_v}{v (\partial p / \partial T)_v} - 1 \right)^{-1}. \quad (10)$$

The utility of Equations (9), which do not involve any assumptions as to the form of the equation of state, resides in the fact that  $\alpha$ , which is properly a function of the state, can be shown to be an essentially positive quantity. Equations (9) with  $\alpha = 0$  thus provide rigorous upper limits to the pressure and density. Furthermore, it can be shown to be plausible that

$$0 < \alpha < 1/4,$$

so that Equations (9) also provide estimates of lower limits to the pressure and density. By applying a previously proposed equation of state, Equation (3), Jones has estimated that  $\alpha = 0.20$  for the case of PETN. This procedure thus makes it possible to estimate the pressure and density (and, by means of the Hugoniot equation, the enthalpy) and also to give a rigorous upper bound and plausible lower bound to this estimate.

### CONCLUSION

A considerable amount of investigation evidently is required before the appropriate form of the equation of state can be unambiguously established. The direct experimental determination of additional properties of the detonation wave would determine the correct form of the equation. Measurement of the temperature would supplement most effectively experimental detonation velocities for this purpose, since the calculated temperature is the most sensitive to the form of the equation of state. It seems probable that measurement of the time dependence of the velocity of unsteady detonation waves would yield valuable supplementary information. Theoretical studies of the energy of gases at high temperatures and very high pressures might serve to establish the proper form of the equation of state.

At present, it is felt that the Kistiakowsky-Wilson equation of state offers the most plausible description of the state of detonation gases. Unfortunately, the parameters of this equation which are now employed are obsolete, since the velocity data employed in their evaluation have been largely superseded by data of greatly improved accuracy. Since progress is likely to be slow in the various different lines of investigation toward elucidation of the proper form of the

<sup>13</sup>J. Hirschfelder, D. Stevenson, and H. Eyring, J. Chem. Phys., **5**, 896 (1937)

<sup>14</sup>H. Jones, Proc. Third Symposium of Combustion, Flame, and Explosion Phenomena, The Williams & Wilkins Co., Baltimore, p. 590 (1949)

equation of state, it is probably desirable that the parameters of this equation be recalculated, using the best available rate data. Because of the formidable numerical nature of these calculations, modern automatic high-speed computational equipment should be employed in this task.

#### DISCUSSION

DR. KIRKWOOD: Dr. Brinkley's paper is now open for discussion.

DR. COOK: Following the suggestion of Dr. Brinkley, I would like to say a few things about this Paterson criticism and also my  $\alpha(v)$  approximation. Actually, I agree thoroughly with one of Paterson's points and with Dr. Brinkley on that particular point. Paterson and I really do (and always did) agree, however, in regard to this  $\alpha(v)$  approximation. I don't know whether any of you are familiar with his work, but he started with a completely independent equation of state which includes the  $\alpha(v)$  approximation; that is, an equation of state obtained by evaluating the parameters theoretically and in this particular  $\alpha(v)$  form. As you know—those of you who are familiar with that work—he was able to calculate detonation properties and to calculate pressures which were in agreement with all of these other methods and velocities which agreed fairly well with experiments. There is one difficulty that I see and that is that his velocity increased a little more rapidly with density than the observed velocities. And his curves crossed over the experimental  $D$  vs.  $\rho$  curve. At about a density of 1.2, it crossed over and it had just a little too high a slope, but when you realize that all the parameters were theoretical, I think Paterson's correlations were very significant. Now, in regard to Dr. Brinkley's statement of Jones' work, I think I agree in some respects with Jones' work but I think the internal energy is going to be negative; the internal pressure is going to be negative at high densities.

Incidentally, I worked out a couple of years ago this same thing that Jones has come up with, but I wasn't resourceful enough at that time to put it out as an upper limit. I realized it was an upper limit, and I have that paper here right now. I never published it. I talked it over with Dr. Kistiakowsky and Dr. Paterson and I had some correspondence on it. I think there are some interesting things about the work of Jones that haven't been presented yet. I would like to show you the calculations of pressures corresponding to that upper limit to compare them to those that we have calculated by other methods, and then I think we can get some information as to the nature of the true equation of state from this, that is, from reasonable considerations bringing in external evidence. For PETN at a density of one, we get 86 kilo atm as the upper limit for the detonation pressure compared with 85 kilo atm obtained from my early work. Also, the upper-limit calculation gives at a density of 1.6 a pressure of 273 kilo atm compared with 225 kilo atm from the earlier work.

These are the values I have calculated from this  $\alpha(v)$  approximation, i.e., the values 85 and 225. The others (86 and 273) are the upper-limit pressures which Jones would calculate, i.e., which I computed from an equivalent equation. This method is identical with Jones', and these are those upper limits. Now, I've been working considerably lately on the solid state, and since Jones' considerations do come from solid-state considerations, I think it is of interest to consider that particular thing from the viewpoint of a familiar potential-energy curve of a molecular crystal. In the solid state, we would expect from the potential-energy curve for a molecular crystal (van der Waals crystal) that at about a density of 1.2 you start to run into a net repulsion instead of attraction. That is, the minimum of the curve comes at this density, and at a greater density your repulsion term exceeds your attractive term in the interaction potentials and I think, then, that this correlation at a density of 1.0 to 1.2 is significant. Also, the fact that one obtains a higher pressure at a density of 1.6 than in the  $\alpha(v)$  approximation and a lower pressure at lower densities is significant. Now, in addition, I calculated a number of other explosives and showed the same situation; in all cases at a density between 1 and 1.2 the upper limit and the  $\alpha(v)$  approximation agree; at a density of 1.6 the upper limit is always much higher, and at the very low densities it is always lower than one computes from the  $\alpha(v)$  approximation. As Dr. Brinkley points out, there are a lot of complicating factors so that it is difficult really to understand what this means, but I feel that it means that we

are passing the  $\alpha(v)$  approximation at about a density of 1 to 1.2. Now the form in which I have this is:

$$\frac{dD}{dv_1} = \frac{D}{v_1} - \frac{D}{2(v_1 - v_2)} + \frac{v_1^2}{2D(v_1 - v_2)} \left\{ \frac{dp_2}{dv_2} + \frac{p_2 - p_1}{v_1 - v_2} \right\} \frac{dv_2}{dv_1}$$

where  $D$  = detonation velocity,  $v$  = specific volume,  $p$  = pressure, subscript 1 refers to conditions ahead of the reaction zone, and subscript 2 indicates those immediately behind the reaction zone. If we were dealing with the  $\alpha(v)$  approximation, I think the last term would correspond with the Chapman-Jouguet condition. That is, when the  $\alpha(v)$  approximation is correct, the last term in this equation is zero because then  $dp_2/dv_2$  is the same as  $(dp_2/dv_2)_s$ . This term would be zero. We can then calculate the pressure from the first two terms on the right side of the equation. This (i.e., the equation resulting by dropping the last term) is the equation which is equivalent to the upper limit, so you get your upper-limit pressure from that first part of the equation. When we consider (1) evidence from calculations of numerous explosives, even those with solid residues, and which we have calculated by the  $\alpha(v)$  approximation, and (2) the further fact that the  $\alpha$ -vs.- $v$  curves are the same curve for all explosives, I still have difficulty in seeing that that doesn't mean this  $\alpha(v)$  approximation is about correct. We have used this same curve, after evaluating it from four explosives, on all types of explosives; and so far as I know, it hasn't failed. Dr. Davis and I worked together on this; he carried out experimental velocity measurements. I know he got great delight out of concocting new types of materials, measuring their velocity, and turning the composition over to me to calculate. We always found agreement. There was no case in which we couldn't calculate the actual velocity within about 200 in./sec from that curve. When you realize the number of different types of explosives that came under our consideration (we have worked out cases with 20 or 30 percent water in explosive mixtures and all sorts of compositions, those with no carbon in them, those with high percentages of  $H_2$ , and various things of that sort) you can look at this  $\alpha(v)$  approximation with consideration.

DR. BRINKLEY: May I make one or two supplementary remarks. In the first place, I am afraid that the result is, of course, negative, but I'm afraid we can't avoid the essential indeterminacy of the theory. I believe that we have to conclude that these things have been a very successful case of curve-fitting and, as such, they have a very high degree of usefulness. Jones' work on the estimation of thermodynamic properties from rate data alone, I think, is a formal statement of the fact that certain of the properties were extremely insensitive to the form of the equation of state employed. Again, those considerations will be extremely useful in a practical sense, but they do not in any way solve the problem of determining the adiabatic temperature of the detonation gases. Yet for the detailed study of the detonation process, and for the study of shock wave associated with the detonation process, it is precisely the adiabatic which we must be able to determine.

DR. PRICE: I would like to make a very brief comment on the upper limit of Jones' value. Dr. Brinkley indicated that this was not absolutely determined at 0.25. If you use the pressure and other parameters computed by Dr. Brinkley, using the Kistiakowsky-Wilson equation of state, you'll find you can obtain alpha values from Jones' expression which are above 0.25. For PETN and TNT they will vary with the loading density of the explosive, and in the case of PETN I think the upper limit was up around 0.36. There are other materials and other explosives for which it is slightly higher, so that I think these values confirm the order of magnitude of alpha but do not necessarily confirm the fact that the upper limit is 0.25.

DR. KISTIAKOWSKY: Since I've already spoiled Von Neumann's contribution to explosions, I would like to destroy another idol. Actually the work was done by R. Halford, but the peculiar (fortunately later revised) rules of NDRC in the very early days required that the names of the official investigators—Kistiakowsky and Wilson—appear on the report.



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## CONVERGENT SHOCK WAVES

Arthur Kantrowitz  
Cornell University

We were first attracted to the possibility of making convergent shock waves by the very high temperatures and pressures which you might easily expect to reach in this way. Now, the problem of implosion is really not a new one. It exists in various military devices, as you know, and an analysis of a shock wave proceeding toward a center has already been made, in particular by Guderley during the last war. His results are shown in Figure 1.

We have checked Guderley's solution by solving the same problem by the method of characteristics, and you see that we obtain essentially the same result. The line at the top of the figure represents the pressure and temperature after the shock wave reflects from the center, after it goes down to a point, and comes out again, and we obtained some points by the method of characteristics—those I have given here.

Well, this by itself might easily be highly academic, except that it seemed to us that it was really possible to produce such things because shock waves apparently have a tendency to assume certain very simple shapes. For example, if you have a shock wave propagating down a tube in the usual shock-tube experiment, you'll find that the shock is very flat. Hornig has informed me that his experiments on reflection from shock waves indicate that a shock wave such as that found in a shock tube is flat to within about four minutes of angle after it has progressed down the tube a little way. This is a rather remarkable phenomenon, and it indicates some stability mechanism operating to maintain this flatness. Now, I might point out that the assumption of a special shape by a wave is something that is only possible for a dissipative wave. If you had a reversible process which tended to assume a special shape, this would really be a violation of the second law, but in the case of a shock wave, we have the possibility of a little organization appearing out of the chaos of the dissipation naturally present in the shock wave. The analysis of this stability can best be done, I think, from the point of view of Mach reflection and Figure 2 shows Mach reflection in a schematic form.

Now the characteristic thing about this is that this Mach shock is curved always. Maybe it is best to use a diagrammatic sketch of a Mach reflection (Figure 3). If the Mach shock is curved thusly, then the angle between the air immediately behind the triple point and the incident air is much smaller than the angle between the wall flow and the incident air, so that

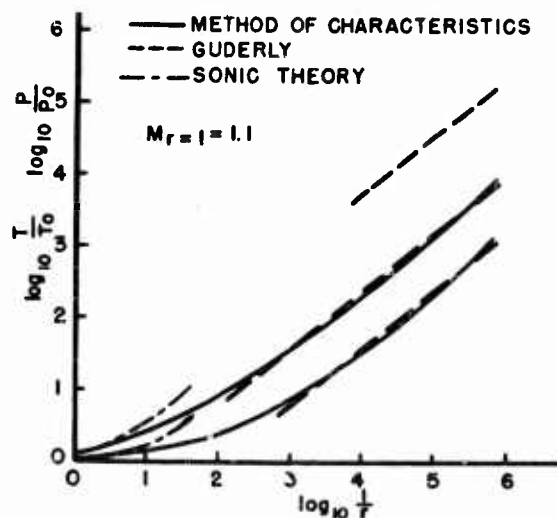


Figure 1 - Comparison between sonic theory, Guderley's solution, and the method of characteristics for spherical converging shock waves ( $\gamma = 1.40$ ). The values  $p/p_0$  and  $T/T_0$  are the ratios of pressure and temperature immediately behind the shock to the initial values. The short upper curves correspond to the pressures after reflection from the center.

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Figure 2 - The entrance of a plane shock wave into a converging two-dimensional channel

when this wave proceeds to the other side and is re-reflected, the re-reflection will be considerably smaller in general than the original reflection. In other words, some of the curvature which was initially concentrated at the corner has been spread out over the Mach shock and this spreading out of curvature is what we term stability of the form.

curvature, and if that equalizes you get a plane. On the other hand, if we have a shock which is moving in a converging channel and the shock must be normal to the two walls, then you get a circular cylinder for the form where the curvature has been equally distributed.

Now you see if you have a shock propagating in a straight tube, we have a boundary condition that the shock must be normal at the two walls. In other words, there must be equal amounts of positive and negative

We have taken the extensive Mach reflection data that was compiled by L. G. Smith at Princeton during the war and plotted up what we call the attenuation of corners. If the attenuation is unity, then it means that a corner disappears in one Mach reflection. If the attenuation is zero, it means the corner propagates out unattenuated and a plot of the attenuation for various incident angles at various Mach numbers is shown in Figure 4. At low Mach numbers you get

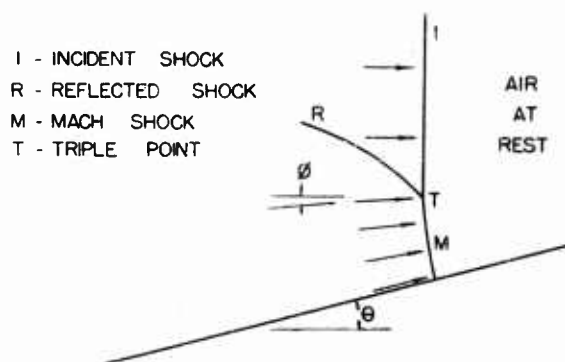


Figure 3 - Sketch of the Mach reflection configuration

very high attenuation in a single Mach reflection, so the corners in a shock wave tend to disappear very rapidly. As you go to higher Mach numbers (Smith data unfortunately gives out at Mach number 2.4) the stability is smaller, and we'll see that this fact plays an important role in what we do from there on.

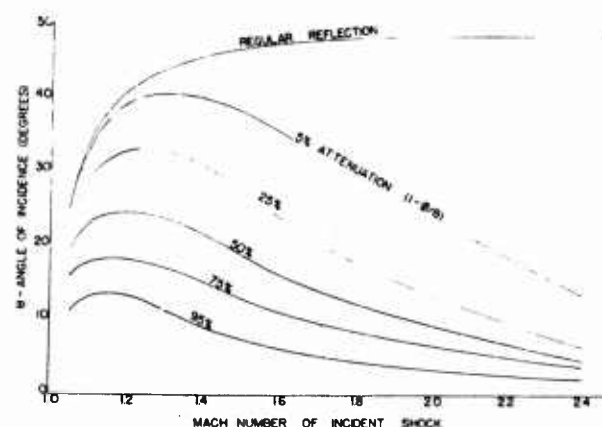


Figure 4 - Attenuation as a function of angle of incidence and Mach number of the incident shock. (From Smith's observations, which were very thinly spaced for the higher Mach numbers of the range shown.)

There is some indication that this rule of a smoothing out of curvatures may not be valid for very high Mach numbers to be found in the linearized theory of Lighthill. Lighthill considers only small corners and he always gets for our attenuation parameter a hundred percent.



In other words, referring to Figure 4, he derives the bottom contour of very small corners but he finds the shock has an inflection point for Mach numbers greater than 2.5 and this would indicate a possibility of instability at high Mach numbers. There really isn't any way of deciding from a linearized theory, and one of the things we are now attempting to do is to work out a second-order theory which would permit calculations of the attenuation of corners in strong shocks. It would be very nice to extend Smith's measurements to strong shocks. Well, this stability that you get is illustrated in Figure 5, where several Mach reflections have occurred in a converging channel, and you see that you get something which is indeed very close to a cylinder. The Mach reflection, even though it looks quite strong, is a very weak shock as you can see from the fact that it does not bend the main shock appreciably.



Figure 5 - Same situation as Figure 2 after the shock wave has propagated some distance into the converging channel

Now to get back to our original interest in this problem, in the attainment of high temperatures, if you continued the convergence of Figure 5 until the walls joined, I don't think you would get anything particularly interesting because at the vertex the heat conduction and the diffusion to the walls would be so high as to dominate the phenomena. We have looked around for other ways of producing a converging shock which would not be dominated by heat conduction and diffusion to the walls. We invented the axially symmetric shock tube that is shown in Figure 6. The shock wave comes up to the point and is pierced by the point and forms an

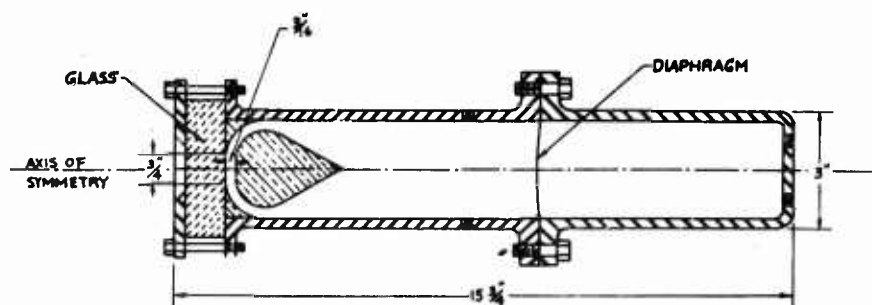


Figure 6 - Sketch of cylindrical shock tube

annulus, and then if it will turn the corner, it will form a converging cylindrical shock wave which has its center of convergence away from surrounding walls. We suspected because of the natural stability of shock waves that, if the shocks were not too strong, it would be possible to produce an angular-ring converging shock wave in this way, but we tested this in a two-dimensional section, and those results are shown in Figure 7.

A cylindrical shock wave is approaching a corner, goes around the corner, and ends up very flat. We built the apparatus shown in Figure 8, and we tried this. There is a glass window to look inside. A camera is in position to take photographs of anything that happens, and the diaphragm flange is also visible.

I would like to present some photographs that we took of the shock wave on the way in and out. We put a mirror on the end of this thing and the Schlieren system—it was a double-up Schlieren system in which the light went in and out again—and in that way we were able to take Schlieren photographs. You'll notice in Figure 9 quite a weak shock; its stability is very

high, and it forms a beautiful circle. It goes right in as far as you can tell, and it comes out and leaves in the center a hot point where the dissipation was very high. As you might expect, it would form a perfect circle on the way out. If you do the same thing again starting with a little stronger shock, you get the results of Figure 10. Here the stability is not nearly so good, and you don't get quite perfect circles. You get a larger heated region, and the heated region is somewhat irregular, showing that the convergence was not perfect.

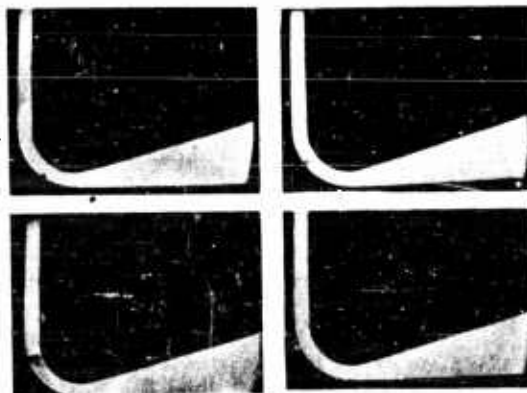


Figure 7 - Schlieren photograph of the progress of a shock wave around a corner in a two-dimensional shock tube

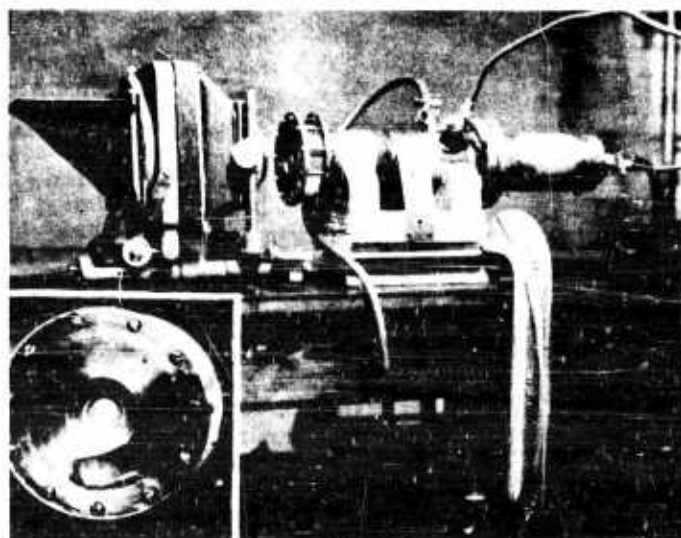


Figure 8 - Original cylindrical shock tube

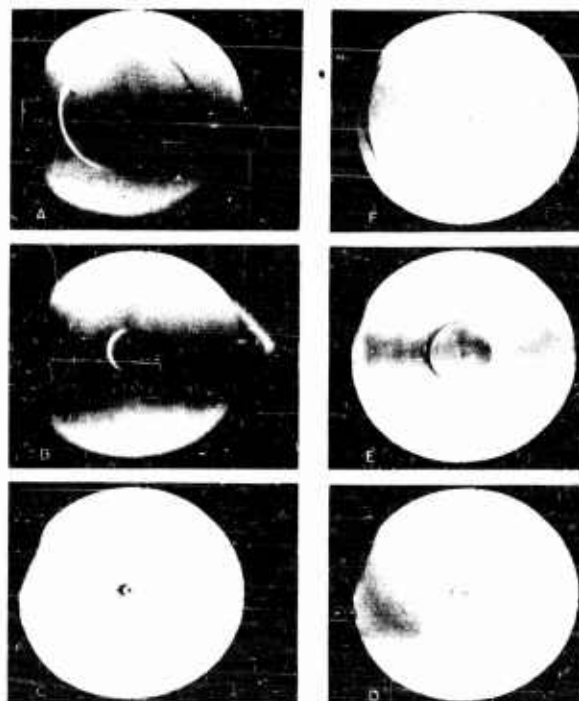


Figure 9 - Schlieren photographs of converging cylindrical shock waves in air (obtained from plane shocks with  $M = 1.1$ ). A, B, C indicate the incident waves and D, E, F indicate the reflected waves. Each photograph is of a different shock wave. The glass window was 1-1/4 inches in diameter

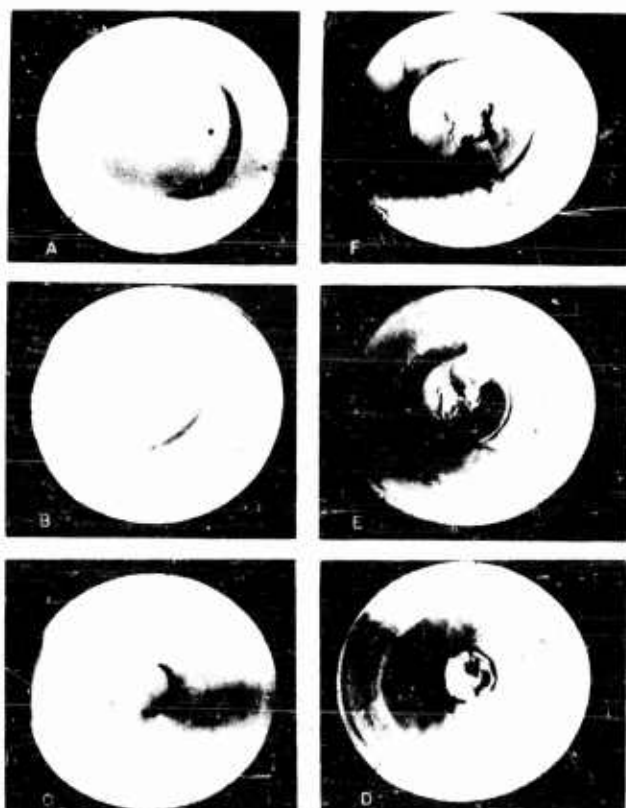


Figure 10 - Schlieren photographs of converging cylindrical shock waves in air (obtained from plane shocks with  $M = 1.8$ ). A, B, C indicate the incident waves and D, E, F indicate the reflected waves. Each photograph is of a different shock wave. The glass window was 1-1/4 inches in diameter

As I said, this convergence drives the shock Mach number up to quite high values. We darkened the room, looked into this shock tube, created a shock with quite a small Mach number, and looked into the glass window at the center. I was willing to bet on our first experiment that you would see a luminous spot. The luminous spots did appear, and we have some photographs of them (Figure 11). This is a three-quarter-inch window, and you see a very tiny luminous spot. Its diameter is limited only by the fact that the optical system we used is quite poor. We don't know how small it is, but it is smaller than a few hundredths of an inch. Now the light that is produced indicates that we are going to at least the temperatures where a certain amount of ionization is present. If you put an electrode some place in this region, you easily establish that the gas becomes conductive for a very short period. Before we could go much further with this type of thing, we really felt that we needed two types of information.

First, we are up against the same kind of problem that has been discussed here this morning of knowing about the state of the gases under these high temperatures and pressures. Well, the pressures aren't high by comparison with what you people are used to, but the temperatures are high. We can use ideal gas laws.

We thought that we could establish that information by experiments easier to interpret than this one. Other information that we need, of course, is about the stability of strong shock waves, but our view

is that the stability will not be high at high Mach numbers. You have to get a good converging shock — get it all smoothed out while the Mach number is low, while it is below two, pressure ratio below five or six — and then let it converge without disturbing it further. We have been doing a little work on the properties of gases at very high temperatures. If you take a garden variety of shock tube with a diaphragm and fill one end with hydrogen and the other end with argon, you can quite easily produce shocks that are strong, even by your standards. For example, if you evacuate this down to one millimeter of pressure and put this in something like ten atmospheres so that you get a 1,000-to-1 pressure ratio, then the theoretical shock which you calculate here is moving at about 4,000 meters per second in a monatomic gas, so it is possible to attain very high temperatures. This gives us an easier method of investigating the properties of gases at high temperatures. First you do a preliminary series of experiments in which you establish that the shock velocity is indeed what you calculate. You'll find it is a little lower than is traditional — ten percent lower or something like that — but its performance is a little different from the low-pressure-ratio shock tube. Then, if you look at the statistical mechanics of argon, you'll calculate that with a pressure ratio you could easily achieve 10,000 to 1. The argon is ionized something like one percent, so it is an excellent conductor of electricity. By that time, theoretically, the gas has other interesting properties that I'll refer

to briefly later. This is all theoretical. Well, we put a little electrode in the end and the electrical conductivity is shown as in Figure 12.

This is just a measure of electrical conductivity, and the early part of this curve indicates the rise in conductivity as the shock wave covered the electrode and the steep rise corresponds to the reflection of shock wave at the end of the tube. Conductivity that we get decays very rapidly and is far less than you calculate from statistical mechanics by perhaps a factor of a hundred thousand. This I don't understand either. We have examined several of the obvious sources of heat loss. Well, first we eliminated by this experiment the possibility that there is a large relaxation time for the appearance of ionization—by the fact that the curve follows what you would expect in its early phases, which means that some ionization appears immediately. The lags are just a few microseconds at most, but the fact that it doesn't build up to its expected values is somewhat mysterious unless there is some large source of heat loss.

One other experiment which we did which I thought might be of some interest was that we took a spectrogram of the light which is attained in such a shock wave (Figure 13). You see that we get a line spectrum. This was just done a few days ago and we haven't had time to analyze this spectrum yet. We get lots more lines than you get from an argon arc with the same exposure, and our immediate intentions are to explore this a little by comparing the spectra of an arc obtained in the shock tube immediately before the shock wave with the light emitted from the shock wave itself to see if there is a real difference when you consider gases of exactly the same composition.

The experiments on shock waves in converging channels were conducted by A. Hertzberg and the author and are reported in the Journal of Applied Physics (Vol. 21, No. 9, 874-878, September 1950). The later converging shock experiments were conducted by R. W. Perry and the author and have been accepted for publication in the Journal of Applied Physics. The most recent group of experiments in straight shock tubes, including the conductivity work and the spectrograms, were undertaken by E. L. Resler, S. H. Bauer, and the author and will be reported more fully shortly.

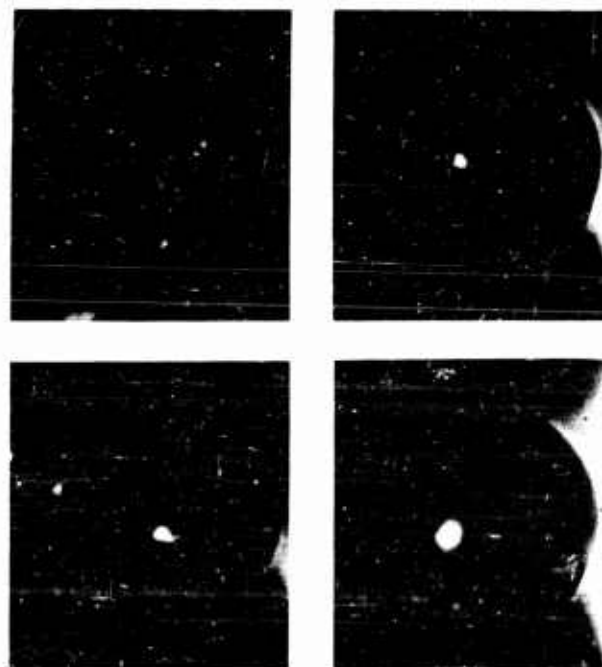


Figure 11 - Luminosity of converging cylindrical shock waves in argon (obtained from plane shocks with  $M = 1.8$ ). The glass window, rendered visible by double exposure, was actually  $3/4$  inch in diameter

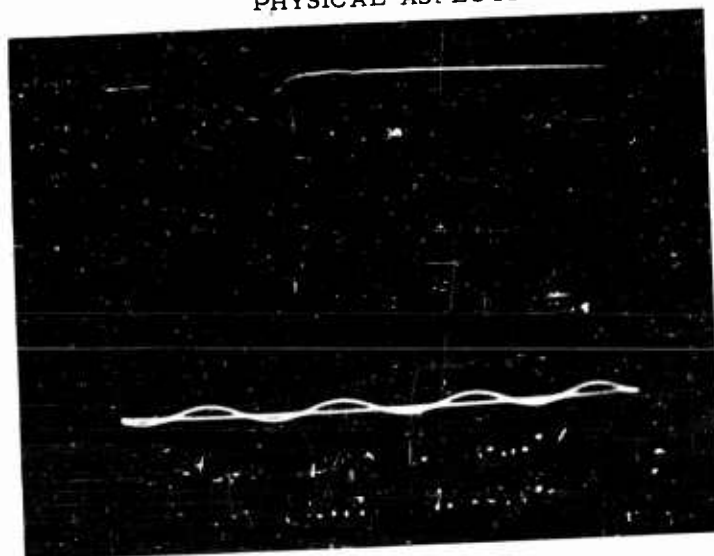


Figure 12 - Oscillogram of the conductivity produced by a shock wave in argon. The low pressure in the argon was 20 cm of Butylphthalate and the high pressure in the hydrogen was 11 atm

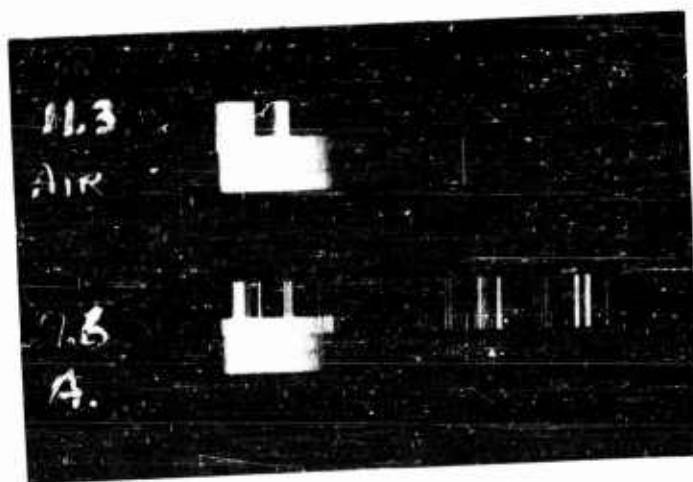


Figure 13 - Spectrograms of the light produced by strong shocks in argon and air. Hg spectra are given for comparison

## DISCUSSION

DR. WEYL: I want to ask what the hopes are of using the shock tube to get information about the equation of state of gases at high temperatures by getting another independent measurement in addition to shock velocity? For instance, density behind the shock front.

DR. KANTROWITZ: One program we have in mind may answer this, and that is to measure the velocity of sound immediately behind the shock. If you know the shock velocity, then the velocity of sound is a nice thing to know. I guess it's about as good as anything. We have done that for low-Mach-number shocks. We intend to do it in ranges where we can get information of interest about the properties of gases. One thing we intend to study in this way is the dissociation of the components of air. Put nitrogen in the shock tube and if you work in a time range, if you adjust the temperature so that the relaxation time is comparable with the time it takes the shock wave to travel across the tube, then you'll see a distortion from the perfect circle which will indicate a subtraction of heat due to the association of nitrogen, so we can measure the association velocity.

DR. LEWIS: I believe Bleakney is measuring the density behind the shock wave in his shock tubes.

DR. KANTROWITZ: I would think that that would be good as long as you have good high densities, but if you were trying to work in the range well below atmospheric pressures, it would be difficult, but it might well be that that could be done. He has quite good precision.

## SHOCK WAVES IN SOLIDS

J. E. Ablard  
Naval Ordnance Laboratory

Dr. Brinkley has given you a discussion of the hydrodynamic-thermodynamic equations of the detonation process and has pointed out the desirability of being able to measure one or more of the quantities independently. One continually meets the difficulty of lack of information on equations of state at high T and P. Calculations of detonation velocities for example or underwater shock-wave parameters soon lead to difficulties because of lack of such information even for the simple gases that appear as the products of detonation.

Dr. Goranson pointed out in 1946 that certain information obtainable from the study of shock waves in solids could supply some of the data. The argument is as follows:

The three hydrodynamic equations applied to the detonation process are:

$$\rho_0 D = \rho (D - u) = m \quad (\text{conservation of mass}) \quad (1)$$

$$mD + P_0 = m (D - u) + P \quad (\text{conservation of momentum}) \quad (2)$$

$$E_0 + mD^2 + \frac{P_0}{\rho_0} = E + m (D - u)^2 + \frac{P}{\rho} - \Delta Q \quad (\text{conservation of energy}) \quad (3)$$

where D = velocity of the detonation front

$\rho$  = density

u = particle velocity

m = mass flow per sec

P = pressure

E = energy

$\Delta Q$  = chemical energy released in the reaction.

The subscript zero refers to the state ahead of the front. Characters with no subscript refer to a certain condition behind the front. In the detonation process there is a steady-state condition defined by the Chapman-Jouguet hypothesis which states that

$$D = u + C, \quad (4)$$

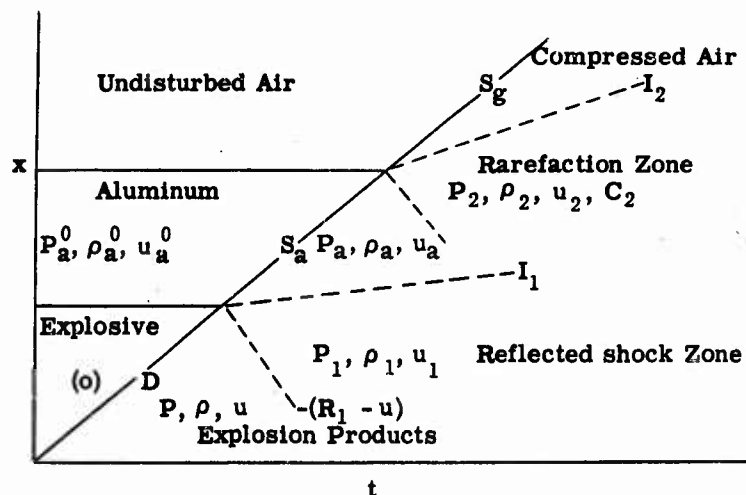
where C is the velocity of sound at  $\rho$ , T, and P. In addition to the above equations, there is an equation of state which for our present purposes is best written

$$E = E(T, \rho). \quad (5)$$

A glance at Equation (1) shows that if the detonation velocity can be measured, and in addition one other quantity, for example, u, the rest of the quantities in Equations (1), (2), and (4) can be calculated.

Let us now review the theory of the transmission of shock waves through a solid, and specifically we will choose aluminum for the solid for reasons which will be apparent later. The shock wave is considered to be generated by a detonation wave in an explosive placed in contact with the aluminum. On an x-t diagram, the situation looks like this:





Analysis of the equations for the detonation wave and the two shock waves, together with the reflected waves and the interface conditions yields the relations:

$$\frac{I_1}{u} = \frac{\rho_0 D + \rho R_1}{\rho_a^0 S_a + \rho R_1} \approx \frac{2\rho_0 D}{\rho_a^0 S_a + \rho_0 D} \quad \text{if } \rho_0 D = \rho R_1$$

$$I_2 = I_1 + \sigma_a,$$

where

$$\sigma_a = \int_{\rho_g}^{\rho_a} \frac{C_2 d\rho}{\rho} = I_1.$$

Hence

$$u = \frac{\rho_a^0 S_a + \rho_0 D}{2\rho_0 D} (I_2 - \sigma_a) = \frac{\rho_a^0 S_a + \rho_0 D}{2\rho_0 D} \cdot \frac{I_2}{2}.$$

These equations have been obtained with the following assumptions:

- (1) that the detonation wave is planar;
- (2) that the shock wave returning into the explosion products has only a small over-pressure;
- (3) that the "escape velocity" of the "free" surface of the aluminum moves with a velocity of  $I_2 = I_1 + \sigma_a = 2I_1$ ,

where

$$\sigma_a = \int_{\rho_g}^{\rho_a} \frac{C_2 d\rho}{\rho};$$

- (4) perturbations due to the effect of a peaked front on the detonation wave have been ignored.

We will return to these approximations shortly.

It is possible to measure  $D$ ,  $S_a$ , and  $I_2$  by an apparatus originally designed by Goranson and altered somewhat to suit our experimental conditions. Briefly, the plane detonation wave is generated in a block of explosive 6x6x5 inches by a plane-wave booster. Aluminum plates of

varying thicknesses from 0.010 inch to 1.5 inches have been used. The plates are drilled to allow insulated probes to be inserted at various known depths. These probes make contact with the bottom of the holes as the shock wave passes through the aluminum. Electrical pulses generated by these contacts are brought through coaxial cables and finally appear on a scope as "pips" on the calibrated sweep of a cathod-ray oscilloscope. From the positions of the pins and the times read from the scope, the shock velocity,  $S_a$ , is calculated. Another set of pins set at various known distances from the "free surface" of the aluminum make contact with it as it moves off into the air. By a similar method of recording and computing, the "free" surface velocity,  $I_2$ , is obtained. In most cases the detonation velocity is inferred from the density of the explosive with reference to the literature, but in a few cases we have been able to measure detonation velocities off the side of the charge by a similar, multiple-contact method. Some results of the measurements are given in Table 1.

TABLE 1

Factors	COMP B	TNT
$\rho_0$ (g/cc)	1.70	1.58
$D$ (m/sec)	7850	6880
$I_2$ (m/sec)	3450	2500
$S_a$ (m/sec)	7250	6880
$U$ (m/sec)	2130	1690
$P$ (kilobars)	283	184
$U$ (calc) (m/sec)	1800*	1500
$P$ (calc) (kilobars)	245*	173
$\rho_0 D \times 10^{-6}$ (gm/sec/cm <sup>2</sup> )	1.33	1.09
$\rho_a^0 S_a \times 10^{-6}$ (gm/sec/cm <sup>2</sup> )	1.96	1.86
$\rho$ (g/cc)	2.32	2.10
$\frac{\rho_a^0 S_a + \rho_0 D}{2\rho_0 D}$	1.237	1.353
$\alpha$	0.23	0.16
$K$	2.68	3.07

\*Extrapolated

In lines 7 and 8 are presented the quantities calculated by Brinkley and Wilson in OSRD 1707. The agreement is certainly qualitatively good and in most cases is within 10 percent.

What contributes to the difference between experimental and calculated values is an interesting field for thought. The calculated values, of course, involve some approximations and assumptions mostly arising, as I have previously mentioned, from lack of data at high temperatures and pressures on the simple gaseous products. The "experimental" values involve the approximations listed above plus, of course, the usual experimental variations. Much has been done to improve the apparatus. Considerably more remains to be done.

However, one would like to be able to estimate the errors incurred as a result of the aforementioned assumptions. Let us return to them.

1. Plane detonation wave. This is an experimental difficulty. Since the pins must be displaced laterally to avoid the effects of one interfering with the measurements from all the others, they are actually placed so that the contacts form a helix. If the wave is not planar to 0.01 microsecond, the velocities measured will be in error. Only a 2-in. diameter region in the center of the charge and plate is used, hence it is believed that this factor is under control.

2. Shock wave returning into the explosion products is negligible. That is,  $R_1 \rho = D \rho_0$

Actually this can be avoided by matching the impedance of the plate and the explosive so that the reflection is reduced to zero as was suggested by Dr. Kirkwood. We expect to accomplish this by use of alloys of aluminum and magnesium. However, it is possible to show that for aluminum this does not introduce an error of more than 1 percent in the particle velocity. If we assume that the Hugoniot curve is approximated by an adiabat of the form  $P = A\rho^\kappa$ ,

it follows that

$$R_1 = \sqrt{\frac{P_1 - P}{\rho \left[ 1 - \left( \frac{P}{P_1} \right)^{1/\kappa} \right]}}$$

Hence for Comp B

$$(\rho R_1)' = 1.41 \times 10^6$$

$$u' = 2120 \text{ m/sec}$$

compared with 2130, which is well within our present experimental error in determining  $u$ .

3. The free surface velocity is twice the particle velocity in the aluminum shock.  
That is,  $I_2 = I_1 + \sigma_a \approx 2I_1$ .

We borrow here heavily from the considerations of "Underwater Explosions," by Dr. Robert Cole, who showed by the application of the Tait equation that  $\sigma$  for water is within 2 percent of  $u$  for pressures up to 80 kilobars. If we assume that the Tait equation is also applicable to aluminum, it is possible to calculate from the present data a value of  $\sigma$ . This is done in the following way:

The Tait equation is:

$$P_2 = B \left[ \left( \frac{\rho_2}{\rho_g} \right)^n - 1 \right].$$

Assuming  $B$ ,  $n$ , and  $\rho_0$  are only functions of the entropy, we readily calculate that

$$\left( \frac{\partial P_2}{\partial \rho_2} \right)_s = C_0^2 \left( \frac{\rho_2}{\rho_g} \right)^{n-1} = C_2^2,$$

where

$$C_0^2 = \frac{Bn}{\rho_g}$$

This allows analytical integration for  $\sigma_a$ :

$$\sigma_a = \int_{\rho_g}^{\rho_a} \frac{C_2 d\rho}{\rho} = \frac{2C_0}{n-1} \left[ \left( \frac{\rho_a}{\rho_g} \right)^{\frac{n-1}{2}} - 1 \right].$$

We were able to fit the Tait equation using  $n = 3$  and  $B = 253$  kilobars. Checking back for  $C_0$ , we find  $C_0 = 5330$ , which compares well with 5400, a value obtained by averaging data from various sources in the literature, including the Lazarus data.

Finally  $\sigma$  was calculated from the integrated equation, resulting in a value of 1730 m/sec, compared to the observed  $I_1/2 = 1725$  m/sec. A further analysis of the situation allowing variance

in  $n$  from 3 to 5 and differences in  $\frac{\rho_a}{\rho_g}$  from 1.3 to 1.5 showed that the maximum error one can expect from the "free surface" assumption is of the order of 3 percent.

4. Perturbations due to the shape of the detonation wave are ignored. I refer here to the VonNeumann picture of the shape of a detonation wave, assuming that the front consists of a shock wave of much higher pressure than at the Chapman-Jouguet condition. The pressure then falls during the subsequent reaction to the Chapman-Jouguet pressure.

Actually, the surface velocity measurements as a function of target thickness have produced a fortunate by-product in the form of confirmation of the essential features of this picture and have allowed estimation of the width of the reaction zone thereby. From measurements on aluminum and other metals, it has been shown that for thin plates the pressure predicted from the surface velocity measurements increases rapidly with decrease in plate thickness to a limiting value some 2.5 times the C-J pressure. Reaction-zone thicknesses were of the right order of magnitude as predicted by Eyring and others. Hence, this experiment shows promise of being a tool to measure the structure of the detonation wave. The picture is fogged, however, by the uncertainty in the above assumption. More work is needed on the theory of the transition of a reactive shock into a pure shock. A recent English publication by Armament Research Establishment, No. 22/50, has thrown additional light on the subject. Conditions of transition from ~~detonation in one explosive to detonation in another are discussed.~~ We have not digested this report completely and do not feel qualified to discuss it at the present time. However, it does not appear to be complete enough to settle our present questions.

What value do these experiments have? Which of the many paths will be the most fruitful to pursue?

If I may return now to equation (3), one will remember that, if we accept the assumptions or eliminate them by some means, we will have a knowledge of all the quantities in equation (3) except  $E - E_0 - \Delta Q$  and that by making measurements at various initial densities we will be able to calculate this quantity at different pressures and densities. The quantity  $E_0$  is the internal energy of the solid explosive at room temperature and one atm. We can conveniently select our standard state such that  $E_0 = 0$ , or it would be possible to calculate the difference between  $E_0$  and the internal energy at any other convenient standard state so that  $E_0$  is a known quantity.  $E$  is the internal energy of the product gases at the C-J pressure, density, and temperature in excess of what they would have at the same standard state. The quantity  $\Delta Q$  is the heat evolved in the reaction if it could be made to take place at the same standard state, and the products must reach the same equilibrium compositions as they would reach at the C-J temperature. Thus, it is here that most of the assumptions in the calculation methods appear. Suffice it to say here that accurate values of  $E - E_0 - \Delta Q$  from the independent surface velocities given as a function of the pressure would permit one to choose between the various assumptions to pick the one which best predicts these additional measurements. As yet we have found it unprofitable to make the calculations because of our uncertainty in  $U$  and  $P$ .

The adiabatic law assumed for the product gases with exponent calculable from such measurements has good possibilities for application in fragment-velocity theory. Since the experiments also give information on the metals, it is now possible to treat the interaction of explosions on metals with more confidence.

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## INTERACTIONS OF DETONATION WAVES WITH MATERIAL BOUNDARIES

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Problems of detonation-wave interactions differ from similar problems involving shock waves in inert materials primarily through the consideration which must be given to the so-called "reaction zone," or the region between the detonation shock front and the Chapman-Jouguet point. The magnitude of this difference is largely determined by the size of this region, and this in turn is primarily a function of the kinetics of the chemical reaction process. Thus, an interpretation of the behavior of detonation waves in nonideal cases would appear to depend in large measure upon methods of analytically introducing the concept of the reaction-zone length into the conventional equations of the thermodynamic-hydrodynamic theory of detonation waves or shock waves.

While there does not at the present time appear to be any universal agreement among workers in the field, either as to the qualitative structure of the detonation wave or as to the general mechanism of chemical breakdown (2, 3, 4, 5), we shall here adopt the point of view previously described by the present authors (2), i.e., the detonation front consists of a shock zone of width possibly some tens of Angstrom units in which abrupt increases of pressure and temperature occur together with a smaller increase of density, followed by a reaction zone in which the heated and compressed explosive reacts to give the equilibrium products at a temperature, pressure, and density as computed by the hydrodynamic-thermodynamic theory at the Chapman-Jouguet point. This reaction zone may vary from 0.1 to some tens of millimeters in length for ordinary condensed explosives. (In explosive dust mixtures reaction zone lengths as great as 10 meters have been reported by (Sir Geoffrey Taylor and others.)

It is convenient in the following analysis to disregard the width of the shock zone itself, together with the complicating effects arising from the lack of equilibrium between external and internal degrees of freedom in this region in an effort to obtain tractable equations. While it is recognized that the Chapman-Jouguet condition strictly applies only to stable plane detonation waves, we shall here effect only a partial correction to this condition following the procedure of Devonshire and others (1, 2), which may be stated as follows: the detonation velocity will be given by the sum of the material velocity and the local velocity of sound, both evaluated at the point in the reaction zone at which reaction is complete, except in the immediate neighborhood of boundaries, in which region these quantities will be evaluated at the point at which the rate of increase of pressure due to the change of chemical composition and entropy just equals rate of decrease of pressure due to the time rate of change of velocity and volume or that due to the curvature of the wave front. It is thus evident that, at least in part, the nonideality of real detonation waves may be ascribed to incomplete release of chemical energy. This effect can be accounted for approximately by assuming the shock front to be curved and such an analysis is used here.

### EFFECT OF CURVATURE ON STABLE DETONATION WAVES

Consider a spherical detonation wave assumed for the moment to be propagating with the stable detonation velocity  $D$ . Then the equation of continuity may be written

$$(D-W) dV/dr = -2VW/r - VdW/dr, \quad (1)$$

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where as usual  $V$  is the specific volume,  $W$  the radial component of the material velocity, and  $r$  is measured radially from the detonation front. Letting the radius of curvature be  $r_0$ , and integrating, one obtains

$$\frac{D-W}{V} = \frac{D}{V_0} + 2 \int_{r_0}^{r_0-a} \frac{W}{Vr} dr, \quad (2)$$

or, defining a quantity,

$$\mathcal{V} = 1 + \frac{2V_0}{D} \int_{r_0}^{r_0-a} \frac{W}{Vr} dr, \quad (3)$$

the continuity equation becomes

$$\frac{D-W}{D} = \frac{V}{V_0} \mathcal{V}, \quad (4)$$

where  $a$  is the reaction-zone length, assumed constant. Using Equations (4) in (3), one may obtain a new form for the equation defining  $\mathcal{V}$ :

$$\mathcal{V} = 1 + 2 \int_{r_0}^{r_0-a} \frac{\frac{V_0}{V} - \mathcal{V}}{r} dr \quad (5)$$

As a first approximation, it may be assumed that the integrand is sensibly constant through the wave, whence  $\mathcal{V}$  takes the form

$$\mathcal{V} = 1 - 2(V_0/V_1 - 1)(a/r_0), \quad (6)$$

which reduces, for a great many solid explosives, to

$$\mathcal{V} = 1 - (a/2r_0) \quad (7)$$

as previously pointed out (2).

The equation for the conservation of energy is, in differential form, the same as it is for the plane wave:

$$dE + d(PV) + UdU = dQ. \quad (8)$$

Integrating, and introducing the Abel equation of state, with a constant covolume  $\alpha$ ,

$$P(V - \alpha) = RT. \quad (9)$$

We obtain then

$$P(\gamma V - \alpha) + \frac{1}{2}(\gamma - 1)(U^2 - D^2) = (\gamma - 1)(\Delta Q + \bar{C}_V T_0). \quad (10)$$

In the above,  $\Delta Q$  is the heat of reaction per gram,  $U$  is the relative material velocity  $D-W$ , and  $\gamma$  is the ratio of heat capacity at constant pressure to that at constant volume.

The equation of motion is again the same as for a plane wave and may be written

$$VdP + UdU = 0. \quad (11)$$

This may be integrated approximately by introducing for  $U$  its value from the continuity Equation (4):

$$dP + (D^2/V_0^2) V d\lambda + (D^2/V_0^2) \lambda^2 dV, \quad (12)$$

which upon integration gives the result

$$P + (D^2/V_0^2) \lambda^2 V + \frac{1}{2} (D^2/V_0^2) \int \lambda^2 dV = \text{Constant}. \quad (13)$$

For not too strongly curved waves, most of the contribution to the integral in Equation (13) will come from values of  $\lambda$  near unity; assigning it this value gives the approximate expression

$$P + (U^2/V) (1 + \lambda^2)/2\lambda^2 = D^2/\tilde{V}_0. \quad (14)$$

In regions far from boundary disturbances, it may be assumed that the Chapman-Jouguet condition may be applied in the same manner as for an ideal plane wave, as discussed above. This leads, as in the traditional equations, to the expression

$$U_1^2 = P_1 V_1^2 \gamma / (V_1 - \alpha), \quad (15)$$

where the subscript 1 refers explicitly to the values of  $P$ ,  $V$  at the point of complete reaction (hence, approximately, at  $r = a$ ). Introducing the value of the ideal (plane wave) detonation velocity,

$$D_1^2 = 2(\Delta Q - \bar{C}_v T_0) (\gamma^2 - 1) / (1 - \alpha/V_0)^2, \quad (16)$$

Equations (4), (10), (13), (14), (15), and (16) lead to an expression of the form

$$\frac{D_1^2}{D^2} = \frac{\gamma + 1}{(1 - a)^2} \frac{2x \left(1 - \frac{x+1}{2} a\right) \left(\gamma^2 + \frac{a}{2}(x-1)\gamma - ax\right) + (\gamma-1) \left(\frac{(x-1)(3x-1)}{4} \gamma^2 + x(2ax^2 - x - 1)\gamma + x^2(a^2 x^2 - 1)\right)}{\left(\frac{1+x}{2} \gamma + x\right)^2} \quad (17)$$

or, for values of  $\lambda$  not too far from unity, the asymptotic expression

$$\frac{D_1^2}{D^2} = 1 + \frac{(2\gamma - 1)a^2 + 3\gamma(\gamma - 1)a + \gamma(\gamma^2 - 2\gamma - 2)}{(\gamma + 1)(1 - a)^2} (1 - \lambda^2) + O(1 - \lambda^2)^2. \quad (17a)$$

For the case in which the reaction-zone length,  $a$ , is comparable in magnitude with the radius of curvature,  $r_0$ , the detonation velocity is not determined by the values of  $W$  and  $C$  (material velocity and local sound speed) at the point of complete reaction but at some previous point—specifically, at the point where the rate of pressure increase due to reaction is equal to the rate of pressure loss due to curvature, that is, the point at which

$$\left(\frac{\partial P}{\partial N}\right)_v \frac{dN}{dt} = \frac{2C^2 W}{V r_0}. \quad (18)$$

Now  $(\partial P/\partial N)_V = P_1$ , and  $dN/dt = 1/\tau$ , where  $\tau$  is defined as the reaction time and is given by the expression

$$\tau = \int \frac{dr}{U} = \frac{V_0}{V_1} \frac{a}{D} \quad (19)$$

Combining these equations gives, in the case that Equation (7) holds, the result

$$(D_1/D)^2 = 3.88 a_1/r_0, \quad (20)$$

with the coefficient 3.88 replaced by a more cumbersome  $f(\lambda, V_0/V_1)$  in the more general case. In view of the approximations resorted to, it has not seemed profitable to use the more exact expression in this instance.

In general, Equations (6) (or (7) if applicable) and (17) determine the reaction-zone length for a given curvature of the wave front and a known detonation velocity. If the wave front is curved, but does not have uniform curvature, we may make the assumption that small regions of the front are approximately spherical and that the equations above are obeyed for such small regions with a given radius of curvature. If the wave as a whole is propagating at a velocity  $D$  and we consider a point on the front whose radius of curvature vector makes an angle  $\phi$  with the axis of the charge, then if the wave proceeds without change in shape the radial component of the detonation velocity is

$$D_r = D \cos \phi, \quad (21)$$

and for a given reaction-zone length the radius of curvature may be obtained from Equation (17) together with the definition of  $\theta$ . ( $D$  of Equation (17) and  $D_r$  of (21) are now identical.) By a step-by-step numerical or graphical procedure, the successive spherical arcs may be connected until the angle  $\phi$  between the normal to the front and the axis reaches  $90^\circ$ , or a value somewhat smaller as determined by the shock-wave velocity in the boundary material. Three cases are considered, depending upon the nature of this surrounding medium.

1. When the explosive charge is surrounded by air or other material of very low density, we may assume that the angle between the attached shock wave in the air and the axis of the charge may be taken as essentially zero. Using the analysis above, with  $0 \leq \phi \leq 90^\circ$ , the values in Table 1 are obtained, where  $R$  is the radius of the charge. The values above

TABLE 1

$a/R$	$D/D_1$
0.00	1.00
0.11	0.95
0.20	0.90
0.29	0.85
0.40	0.80
0.52	0.75
0.68	0.70
0.82	0.65

are appropriate to the detonation of TNT of loading density 1.57 g/cc, covolume  $\alpha = 0.422$  cc/g, and  $\gamma = 1.24$  for the product gases. Similar calculations for other solid explosives indicate that for moderate deviations from ideality one may use the approximate relation

$$(D/D_1) = 1 - A a/R, \quad (22)$$

where  $A$  is a function of the parameters above but differs only slightly from the numerical value 0.5.

2. If we assume that the explosive charge of radius  $R$  is surrounded by a sheath of material of negligible thickness but of mass  $\sigma$  per unit area, then it may be assumed that the confining action arises from the inertia of the case. Thus the limiting value of the angle  $\phi$  will be determined by the fact that the elements of the sheathing move with the same velocity as the adjacent particles of explosive in the direction of the normal to the wave front at the point of intersection. Assuming the pressure at this point, approximately  $P_1$ , the value at



the end of the reaction zone, to be acting for a time  $\tau$ , the reaction time, to produce a momentum  $\sigma W_1 \sin \phi$ , one obtains the expression

$$P_1 \tau = \sigma W_1 \sin \phi. \quad (23)$$

Using ideal wave values for  $P_1$  and  $W_1$ , Equation (19) for the reaction time  $\tau$ , and the definition

$$W_c/W_e = \frac{2\pi R\sigma}{\pi R^2/V_0} \quad (24)$$

for the ratio of the mass per unit length of casing to that of the explosive, one obtains the expression

$$\sin \phi = 2 \frac{V_0}{V_1} \frac{a/R}{W_c/W_e} \quad (25)$$

Equation (25) defines the value of  $\phi$  at which the shock front reaches the edge of the charge in the procedure sketched in case 1 and hence the value of  $R$ , the radius of the charge. Representative values of the quantity  $(a/R)^2/(W_c/W_e)$  are listed in Table 2 for assumed values of  $D/D_1$ , for TNT with properties corresponding to those of Table 1. Empirically, the calculations for a large number of solid explosives indicate that the relationship between  $D/D_1$ , charge diameter  $R$ , reaction-zone length  $a$ , and the relative mass of the charge sheathing  $W_c/W_e$  can be expressed by the equation

$$D/D_1 = 1 - 2.17 \frac{(a/R)^2}{(W_c/W_e)}, \quad (26)$$

where, as before, the numeric 2.17 is an approximation to a function of  $V_0$ ,  $\alpha$ , and  $\gamma$ , and has been averaged over a number of substances. It should also be pointed out that the diminution of the detonation velocity for a given  $a/R$  ratio is only slightly dependent upon the angle  $\phi$ .

TABLE 2

$\sin \phi$	$(a/R)^2/W_c W_e$		
	$D/D_1 = 0.95$	$D/D_1 = 0.90$	$D/D_1 = 0.85$
0.065	0.017		
0.075	0.017		
0.10	0.020	0.041	
0.15	0.020	0.042	0.066
0.12	0.020	0.043	0.066
0.25	0.020	0.044	0.068
0.30	0.021	0.045	0.071
0.35	0.022	0.045	0.070
0.40	0.024	0.047	0.073
0.45	0.024	0.050	0.076

3. If, on the other hand, the bounding material is effectively of infinite extent but of considerable density relative to air, it may be assumed that interaction of the shock front with the surrounding medium is due to the setting up of a shock wave in this material. Here we must assume that the pressure and the particle velocity agree at the point of intersection of the detonation shock front and the material boundary. We assume as before that the normal component of the detonation velocity at the intersection is  $D_r$ , and that the normal to the shock front makes the angle  $\phi$  with the axis of the charge. The shock wave in the casing has the normal velocity  $D_c$  at a corresponding angle  $\phi_c$ . That is,

$$D_r = D \cos \phi_r, \quad D_c = D \cos \phi_c. \quad (27)$$

Matching pressures and material velocities at the boundary leads to the condition

$$P_c = P_r, W_c \sin \phi_c = W_r \sin \phi_r. \quad (28)$$

The hydrodynamic conditions in the explosive and in the surrounding material respectively are

$$P_r V_0 = W_r D_r, P_c V_c = W_c D_c. \quad (29)$$

Combining Equations (27), (28), and (29), one obtains for the angle  $\phi$  the expression

$$\tan \phi = (V_{0c}/V_0) \sqrt{(D^2/D_c^2) - 1} \quad (30)$$

Since the velocity  $D_c$  of the attached shock wave in the case is seldom known experimentally, it must be calculated from the sonic velocity  $C$  in the surrounding material and its equation of state; for velocities not too far from sonic, the velocity of the shock wave in a solid may be expressed as

$$D_c = C (1 + AP), \quad (31)$$

where  $A$  is the compressibility of the solid. Since the pressure is given in the present approximation as

$$P = \frac{D^2 \cos^2 \phi}{V_0} \frac{1 - a}{1 - \gamma}, \quad (32)$$

it is possible to determine  $\phi$  from the preceding three equations. Again returning to the procedure of case 1, knowledge of  $\phi$ , the angle with which the normal to the wave at the point of contact with the boundary meets the axis of the charge, permits the calculation of the reaction-zone length,  $a$ , for any given value of the ratio  $D/D_i$ , the deviation of the actual detonation velocity from the ideal value. Table 3 gives calculated results for a typical case (TNT with constants as before). Again it is possible to average the results for a variety of substances and write

$$D/D_i = 1 - 0.88 (a/R) \sin \phi, \quad (33)$$

where the angle  $\phi$  must now be determined from the characteristics of the casing material.

Calculations based upon Equations (22), (26), and (33) give, for the reaction-zone length,  $a$ , the values given in Table 4.

TABLE 3

$\sin \phi$	$(a/R) \sin \phi$ when $D/D_i =$		
	0.95	0.90	0.85
0.04	0.053		
0.05	0.045		
0.065	0.042		
0.075	0.043		
0.10	0.050	0.109	
0.16	0.050	0.104	0.165
0.20	0.048	0.107	0.164
0.25	0.051	0.111	0.171
0.30	0.052	0.112	0.178
0.35	0.054	0.112	0.175
0.40	0.059	0.118	0.183
0.45	0.061	0.124	0.190

TABLE 4

Substance	Confinement	Loading Density	Reaction Zone (cm)
<sup>1</sup> Picric Acid	Glass	0.90 g/cc	0.22
<sup>2</sup> RDX	Glass	0.90	0.083
<sup>2</sup> 60/40 Amatol	Steel	1.48	0.398
<sup>3</sup> Minol-2*	Lead	1.69	0.538
<sup>3</sup> TNT**	Steel	1.613	0.0346
<sup>3</sup> TNT**	Steel	1.60	0.036

<sup>1</sup>Parisot, A., and Laffitte, P., Congr. chim. ind., Compt. rend. 18me congr., Nancy (1938), 930-6

<sup>2</sup>Copp, J. L., and others, Unpublished reports

<sup>3</sup>MacDougall, D. P., and others, Unpublished reports

\*Angle  $\phi$  taken as  $20^\circ$  from the velocity in lead of a shock produced by Pentolite, 2745 m/sec

\*\*Angle  $\phi$  taken as  $8^\circ 10'$  from the velocity of sound in steel, 5130 m/sec

#### TIME-DEPENDENT DETONATION WAVES

Let us suppose now that a stable detonation wave, either ideal or otherwise, passes from one material into another, where the second medium is characterized by different physical and chemical properties, in particular by a different density and heat of reaction. During the period between one stable regime and the next, an unstable time-dependent detonation wave will exist which may be characterized analytically either by its change in reaction-zone length or by a change in the curvature of the wave front with time. Since it has not been found possible to take into account these factors explicitly, a somewhat different approach has here been followed. It is known that the detonation velocity is determined by the sum  $W_1 - C_1$  at the end of the reaction zone. For a wave whose velocity is changing, the conditions at the Chapman-Jouguet point will not be apparent at the shock front for a time  $\tau$ , where  $\tau$  is the time required for a signal to traverse the reaction zone and is essentially the reaction time. Thus, if

$$C_1 - W_1 = D\tau, \quad (34)$$

and if, for small perturbations, we may write

$$D\tau = D + \tau (dD/dt), \quad (35)$$

we obtain the expression

$$(dD/dt) = -D - W_1 - C_1 = -U_1 - C_1. \quad (36)$$

Again, for small perturbations, it may be assumed as a first approximation that the equation of continuity and that for the conservation of energy apply as for a plane wave, without change, whence

$$U_1/D = V_1/V_0 = (\gamma + \alpha/V_0)/(\gamma + 1) \quad (37)$$

$$P_1(\gamma V_1 - \alpha) + \frac{1}{2}(\gamma - 1)(U_1^2 - D^2) = (\Delta Q + \bar{C}_v T_0)(\gamma - 1) \quad (38)$$

and, introducing the definition of the velocity of sound, and changing the time derivative to a space derivative,

$$C_1^2 = P_1 V_1^2 \gamma / (V_1 - \alpha) \quad (39)$$

$$\tau (dD/dt) = a(V_0/V_1) dD/dx. \quad (40)$$

One finally obtains the result

$$a(dD/dt) = (V_1/V_0)^2 \left\{ \sqrt{D_i^2 - G(\alpha, \gamma, V_0)(D_i^2 - D^2)} - D \right\}, \quad (41)$$

which, for small  $D_i - D$ , may be approximated by the expression

$$a(dD/dx) = 0.333(D_i - D), \quad (42)$$

where the coefficient 0.333 is again a mean value for solid explosives. This approximately logarithmic decay equation has been verified for a few detonation waves decaying from an initially high velocity. It should be pointed out that the quantity  $D_i$  in Equations (41) and (42)—the ideal velocity—may be replaced, in the present approximation, by any stable velocity related to the ideal, plane-wave velocity by the preceding equations. The scattered results presently available are assembled in Table 5.

TABLE 5

<sup>1</sup> 2C0 + O <sub>2</sub> , $D_i = 1.76 \times 10^5$ cm/sec	$a = 1.1$ cm
<sup>2</sup> Nitroguanidine, $V_0 = 1.89$ cc/g	0.88
<sup>3</sup> TNT, $V_0 = 1.0$ cc/g, $R = 1.27$ cm	0.076
<sup>4</sup> 80/20 Amatol, $V_0 = 0.61$ cc/g	3.9
<sup>5</sup> Ballistite, $V_0 = 0.954$ cc/g	0.22

<sup>1</sup>Bone, W. A., Frazer, R. P., and Wheeler, W. H.,  
Phil. Trans. 235, 29-68 (1935)

<sup>2</sup>Photograph number 3141, ERL, Bruceton, Penna.

<sup>3</sup>Photograph number 11, ERL, Bruceton, Penna.

<sup>4</sup>Photograph number 3737, ERL, Bruceton, Penna.

<sup>5</sup>Gurton, O. A., Proc. Roy. Soc., A204, 32 (1950)

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## DISCUSSION

DR. KIRKWOOD: Dr. Parlin's paper is open for discussion.

DR. JACOBS: In connection with the curvature of the wave front, I wanted to make a couple of comments. When Jones proposed his theory, the expansion of the gases in back of the wave front, he said nothing about the shape of the wave front itself, but he used a "Meyer" approximation for the outflow; that is, the flow around a wedge. That is a fairly good approximation, and actually our present knowledge on rarefaction theory gives us a chance to look further into that. I don't think anyone has ever done too much about trying to draw a characteristic net through the reaction zone, but there has been some work done (by the Germans on wedges) of drawing a characteristic net behind a detonation shock. Actually there is a bibliography reference to it in Courant and Friedrich's excellent book on supersonic flow and shock waves. The paper has been translated by Brown University recently. I think it's of interest.

DR. PARLIN: I think it is a useful approximation to assume this region is quite small, and I think this can be small for the wave shapes that people have reported in literature and photographic studies.

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SESSION IV

12 January 1951

PROBLEMS AND FUTURE DEVELOPMENTS

George B. Kistiakowsky  
Harvard University  
Chairman

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## PROBLEMS AND FUTURE DEVELOPMENTS

George B. Kistiakowsky  
Harvard University

We started this meeting with remarks by the first chairman on how nice it was to see so many old friends. I'd like to say that after two days it's still nice to see them all here. But at the same time this feeling is slightly moderated by another feeling—that it would have been nice if there were more new faces. I have, somehow, a feeling that maybe the rate of increase of personnel, of scientists who are interested more or less in the field of detonation and explosion, is not quite keeping pace with the needs of this country. And I hope that Dr. Roberts, later on, will talk a little more on this subject.

I have another topic which I will take a few minutes to discuss, and that is in reference to the Zeldovich-Von Neumann priority claim. This fall, having a little more time than I had for several years, I decided it was a good time for me to read some Russian literature in the original, since I am able, with a little difficulty, to do so. And having read, now, about a dozen papers, I got a very uncomfortable feeling that we do not have that order of magnitude of technical superiority over the Russians, at least in the field of explosions, that the newspapers almost convinced me we have generally.

And to give you just an example, here is the Von Neumann discussion of the structure of detonation waves, which was classified, and was terribly important in the development of our thinking. But Zeldovich publishes the same thing in open Russian literature three years earlier. That same paper by Zeldovich contains a reasonably adequate treatment of the charge diameter effect on the detonation velocity as applied to gases—again, something like two years ahead of Jones' discussion applicable to solid explosives.

Once they have the gas theory, there remains such an obvious extension to the solid explosives that one can have no doubt but that they had a full understanding of these phenomena sooner than we had.

During the war, G. I. Taylor developed what quite a few of you have heard referred to as the Taylor instability—what happens when a medium of low density pushes a medium of high density (which is, for instance, applicable, to the expansion of a gas bubble in water). This, again, had rather important, at least military-scientific, applications; but Landau has published the same discussion applied to gases in open Russian literature in '43—a little bit ahead of Taylor's classified report.

That is not all, because, for instance, in '46 I ran across a paper in which delays in initiation of lead azide, measured in fractions of microseconds, were discussed under special conditions. And you see, again in '46 something was published that we're very much interested in right now.

So I'm quite concerned about this and quite interested in dispelling the notion that we are automatically ahead of them. And it seems to me we have to realize that we are not—certainly not in this field—and that we have to make considerable effort to stay ahead, if we are ahead, or to forge ahead if we aren't.

Incidentally, as one minor corollary to that feeling, it seems to me that it would be highly desirable if this Russian literature—which, of course, since it is open literature, largely

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concerns gaseous explosions but does contain a lot of interesting thinking applicable generally—were translated into English by competent translators. I've seen one or two translations done by NACA, and at least one of those is completely unintelligible. The man who did the translation knew the English and knew the Russian, but didn't know either kinetics or hydrodynamics.

Well, I hope that ONR or some powers that be elsewhere can take some steps in this direction. It represents the outlay of some money, and finding the right personnel—among whom I'm afraid I can't be counted—to do the job. Well, my time is limited, and I won't steal all the fire from Jack Kirkwood's coming remarks on new developments, but I couldn't resist saying a few things about this general point. After having listened to the discussions, I feel that very interesting and very significant progress is being made but that we don't know everything there is to know about detonations and related scientific areas. And I am talking not from the point of view of military hardware but that of general understanding of the phenomena involved. For instance, we ought to just mention the equation of state, which certainly has been discussed, but which certainly is not settled.

The question of solid residue from an equation-of-state point of view has not been exhaustively treated, and so there are, I think, very important and interesting and challenging problems in the general field of shocks, which are tied in to the effectiveness of explosives—for instance, the impact-loading of solid materials and also shocks in liquid and gaseous media.

I think that the initiation mechanism, thought of broadly, is extraordinarily important. I mean, already as a question of hazards in handling munitions, it has an important practical application which some of us, at least, do know. There were unpleasant catastrophes during the last war which should not have happened if explosives had only the sensitivity which they were supposed to have. In other words, occasionally they seem to have very different sensitivity.

Questions like that have not received really scientific explanation. One thing that occurred to me, of really quite considerable interest, as I was listening to the talks in this conference, was the observations on hydrazine nitrate, where we notice an extraordinary drop in velocity at high density. It was also pointed out that amitcl levels off in velocity at high densities. Velocity does not increase with density as it should, according to the normal equation of state. In some dynamites, as I recall, there were very definite maximums observed, just as they were observed in hydrazine nitrate.

Now, it's obviously tied in with the thickness of the reaction zone, because the effect seems to vanish in cartridges of large diameter—or at least is minimized. And that, in turn, suggests that the voids between the solid particles in an explosive have some very important role in propagating detonation. That has never been really sufficiently studied and understood. We know perfectly well that we can make explosives extraordinarily insensitive to detonation propagation by bringing the density nearer the true crystal density. Hence, as we take out the voids from the explosives, it becomes more and more difficult to propagate detonation.

I won't mention, now, the more applied work, but I feel that in that area, also, a great deal can be undertaken. But the problems I mentioned—at least, a good fraction of them—can be undertaken not only in government-organized and—managed laboratories, but even, some of these problems, in colleges and small private laboratories. The more applied work, under present conditions, probably has to be limited to government establishments.



## THEORETICAL DEVELOPMENTS IN DETONATION

John G. Kirkwood  
California Institute of Technology

Substantial advances in our theoretical knowledge of the process of detonation in high explosives were made during World War II. New general theoretical approaches were suggested by G. I. Taylor and J. Von Neumann. The structure of the reaction zone was studied in detail by H. Jones and by H. Eyring on the basis of the nozzle theory and the curved front theory, respectively. Systematic calculations of detonation velocities, based on the Hugoniot conditions, the Chapman-Jouguet condition, and empirical equations of state were carried out by H. Jones, by E. B. Wilson, Jr., and by S. R. Brinkley, Jr. In spite of these notable advances, which were of great practical importance in guiding experimental investigations and in providing a basis for decisions on the military use of high explosives, many fundamental aspects of detonation phenomena remain obscure and are in need of further investigation.

Among the general problems requiring further investigation are those of initiation and the propagation of nonsteady detonation waves in solid and gaseous explosives. For this purpose a more detailed study of the structure of the Von Neumann spike, in which the detonation reaction takes place, and of its relation to the Chapman-Jouguet condition, seems to be called for. The dynamical investigations of the Chapman-Jouguet condition by G. I. Taylor and by J. G. Kirkwood and S. R. Brinkley, Jr., provide a starting point for these studies. Such studies should throw light on the mechanism of the transition from deflagration to detonation, which is at present only very incompletely understood.

In the exploration of new explosives of increased power, studies of the kinetics of the detonation reaction with solid reactants, of which aluminum is the representative example, seem to be essential. For underwater use of such new explosives, it is of critical importance to know the distribution of energy between the shock wave and bubble oscillations. This problem should receive special attention both from the theoretical and experimental points of view. It is related not only to the detonation process in the explosive but also to the propagation of the explosion wave in the exterior medium, air or water, in which the weapon is used. For rapid approximate estimation, the propagation theories of J. G. Kirkwood and S. R. Brinkley, Jr., can be used. For an understanding of the finer details of the transmission of the energy of a detonation wave into the exterior medium, more exact integrations of the equations of hydrodynamics with the use of modern computing facilities seem to be called for.

Although the theoretical prediction of detonation velocities has met with considerable success, it frequently fails for explosives leading to products outside of the composition range of conventional explosives. These failures are unquestionably due to inadequate knowledge of the equation of state of the product mixtures. In order to make these aspects of the theory of detonation reliable, a fundamental study of the equation of state of fluid mixtures in the range of thermodynamic variables encountered in explosion products is imperative. On the theoretical side, the best compromise would seem to be to utilize the free-volume theory of Lennard-Jones and Devonshire to the fullest extent, in spite of the approximate nature of this theory. For this purpose it would be necessary to extend the free-volume theory to gas mixtures. Some progress along these lines has already been made by I. Prigogine. On the experimental side the most promising approach appears to be the study of P-V-T relations in gas mixtures, the composition of which simulates that of explosion products, by transient techniques, for example with the pin technique for measurement of particle velocity and techniques involving the observation of the propagation of shock or rarefaction waves in fluid mixtures.

I will conclude my remarks by elaborating a bit on equation-of-state problems. As Dr. Brinkley pointed out this morning, it is impossible to determine the equation of state of the products of an explosion from a study of detonation velocity as a function of loading density. This was not at first recognized, but it is responsible for the fact that so many different empirical equations of state yielded satisfactory results in representing this particular relationship.

Of course, it is possible to determine the equation of state, using the detonation process as an experimental technique, if one measures certain other variables—for example, particle velocity, which is difficult to measure, or temperature, which is still more difficult. But these approaches to the equation-of-state problem are essentially experimental.

Dr. Brinkley and Dr. H. Jones have proved—Dr. Brinkley outlined the proof this morning—that the equation of state is not determined by the information which one gets merely from studying detonation velocity as a function of loading density. And from the theoretical point of view, it seems rather futile for this reason to explore other empirical equations of state. A number have been used, and used with a certain amount of success. Rather, it would seem desirable to do the best one can with a purely theoretical equation of state. As I have mentioned in my remarks, I believe that the Lennard-Jones-Devonshire equation of state, based on the free-volume theory, provides the best compromise. It is sufficiently simple so that the computations will not be hopelessly involved, and I believe that it is sufficiently accurate from the range of thermodynamic variables encountered in explosion products to give reliable results.

A recent survey of the applicability of the Lennard-Jones-Devonshire equation of state to pure fluids—pure gases and liquids—has been made by Dr. Hirschfelder. He finds that the equation of state is very poor at intermediate densities—densities of the order of the critical density or less—but that at densities of the order of ordinary liquid densities the equation of state is quite reliable.

I could outline certain of the quantitative features of setting up a free-volume theory for mixtures, but I think I will not take our time to do this. I will merely mention some of the assumptions which can be made. One will make the usual assumptions of the free-volume theory for pure liquids, and, in addition, one will assume that in the detonation front the neighbors of any given molecule are randomly distributed, that is, that the fraction of nearest neighbors of a given molecule in a liquid of a given type will be proportional to the gross mole fraction of molecules of that type.

Combining this hypothesis of randomness with the other features of the free-volume theory, one can obtain an equation of state for fluid mixtures which is not especially complicated and which I believe may settle, once and for all, when the appropriate calculations are made, the controversy about what equation of state should be used for explosion products and should allow, I think, for the extension of calculations of detonation parameters with some degree of confidence to explosives yielding products of unconventional composition. And, after all, that is what is necessary if the theory is to be fruitful.

#### DISCUSSION

DR. SNAY: I will make a short remark on the evaluation of the covolume factors of the Kistiakowsky-Wilson equation of state which we have made at NOL. Most of the failures in the prediction of the detonation phenomena mentioned today are probably not due to any inadequacy of this equation of state but to the fact that its covolume factors were incorrect. For instance, the covolume factor for  $H_2O$  was believed to be of the order of magnitude of one hundred. In our investigations this factor was found to be near to the value two hundred and fifty. As long as such discrepancies exist, it is not surprising that unsatisfactory results are obtained. It should be mentioned that this procedure is a straightforward determination of the empirical parameters of an assumed equation of state from measured detonation rates, and it

is interesting to note that the values recently obtained are very close to those first published by Kistiakowsky and Wilson in 1941.

As far as the evaluation of the covolume factors is concerned, it is true that this should be done with the use of equilibrium calculations—as pointed out by Dr. Brinkley in his paper today. We did not apply equilibrium calculations but performed the evaluation for two typical product compositions. First, we assumed that all the available hydrogen is completely oxidized to  $H_2O$ . The oxygen left is assumed to combine with the carbon forming  $CO_2$  and  $CO$  in oxygen-rich explosives or  $CO$  and solid carbon in oxygen-deficient explosives. In the other set of calculations it was assumed that  $CO$  is formed first of all. (All this applies to nonaluminized explosives.) These two arbitrary modes of the decomposition of an explosive include a fairly wide range of possible product compositions. The surprising result was that we obtained practically the same covolume factors in both cases. We might, therefore, conclude that the determination of the covolume factors is not too sensitive to the assumed composition of the products.

MR. RUTH: Professor Kirkwood mentioned several problems that still need attention. In that connection, I would like to mention that the matter of sensitivity is still one of the most poorly defined, or at least poorly understood, of the aspects of explosions. And it seems to me that it would be profitable, as the theory of detonation is developed, to attempt to establish a theoretical relationship between sensitivity and detonation.

For example, if we define sensitivity as the ability to propagate the detonation wave stably, we might, by measuring the rate of detonation in two columns—one large and one small—be able to calculate the minimum diameter of a column of a particular explosive which would give stable detonation and use this minimum diameter as a criterion of sensitivity. Following along these lines, we might eventually establish some sort of an absolute criterion on sensitivity.

MR. STRESAU: I might suggest that sensitivity is a more complicated problem than that for the simple reason that, in handling, deflagration might be almost as disastrous as detonation. And we definitely know that some materials, which are rather easy to ignite, are rather difficult to detonate. Materials which are more readily detonated and which are harder to ignite are also well known. For example, in high explosives, PETN is a good bit easier to detonate than tetryl, yet a good bit more difficult to ignite.

Take lead azide and lead styphnate, for example. Lead styphnate is very sensitive to static. It's very easy to get enough of a charge on your body to set off lead styphnate. I've seen it demonstrated very nicely. Lead azide takes much more energy to set it off. But still, lead azide will detonate almost immediately and will detonate in very small columns, whereas lead styphnate is a very difficult material to get to detonate at a high rate.

This is an example of the difficulty in that approach. The difficulty is that sensitivity is so many things it's hard to get any kind of an absolute criterion of sensitivity. This, I imagine, is the big difficulty with sensitivity, anyway—that it's a combination of so many factors.

This, of course, brings up the question of the transition from deflagration to detonation. Perhaps this is a problem that can be expressed in terms of a general relationship which applies to both processes. The rate of propagation of the reaction of an explosive material, whether it is detonation or deflagration, is always a direct function of pressure. We usually consider either stable detonation, which involves very high pressures and velocities, or stable deflagration, which involves much lower pressures and velocities. It is reasonable to believe that these processes are related by a continuous curve in the pressure-mass rate plane. The two processes may not be as different as is usually supposed. As the pressure is increased, a point is reached where it is difficult to determine whether the velocity gradient is due to the improvement in heat conductivity with increasing pressure or the increase in shock velocity with pressure. This point of view is supported by the fact that, with various primary explosives

under various conditions, we have observed propagation velocities covering nearly the whole velocity spectrum. Thus the point at which deflagration turns to detonation may be rather hard to define.

The transition from deflagration to detonation is usually so sudden because the intermediate reactions are generally extremely unstable. This instability can be traced to rather simple considerations.

In the reaction of any explosive we must consider a system, not unlike that of a solid-propellant rocket, in which both the rate of gas evolution and the rate of gas escape are direct functions of pressure. The rate of gas evolution is directly related to the propagation rate. (In a column which is reacting at the end only, the rate of gas evolution is proportional to the propagation rate.) The velocity in such a system is stable only if the rate of evolution is equal to the rate of escape, and the rate of escape increases more sharply with pressure than does the rate of evolution. If the rate of evolution is less than the rate of escape, the pressure, and hence the propagation rate, will reduce to such an equilibrium point or extinguish. If the rate of evolution exceeds the rate of escape, the propagation accelerates until stable equilibrium is achieved.

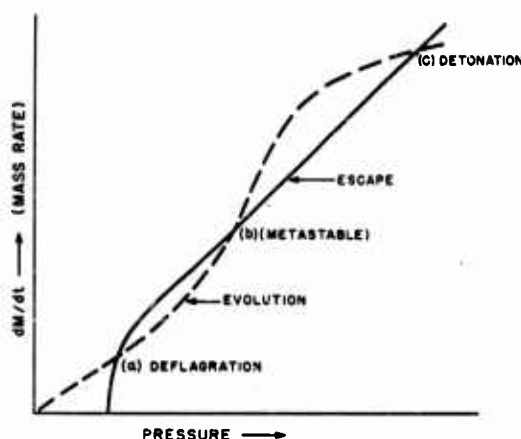


Figure 1 - Mass rates of gas escape and evolution vs. pressure

These ideas are expressed graphically in Figure 1, where  $dM/dt$  is the mass rate for both evolution and escape of the gas and  $P$  is pressure. The solid curve is the escape velocity versus pressure, and the dotted curve is the rate of evolution versus pressure. The crossings at (a) and (c) are stable equilibria of the kind mentioned above, and that at (b) is a metastable equilibrium above which the reaction will accelerate to (c) and below which it will die out to (a). For the purpose of the present discussion, the shape of the curves is unimportant, except that the three crossings are necessary. The two crossings at (a) and at (c), which correspond to stable processes, are necessary since at least two stable processes, detonation and deflagration, are observable for almost all explosive materials. The crossing at (b) is a necessary condition for the other two.

In general, the shape of the pressure-escape rate curve is determined by the hydrodynamic properties of the surrounding system, while that of the pressure-evolution rate curve is determined by the properties and state of aggregation of the explosive.

All rate phenomena connected with explosive reactions can be described in terms of these curves. In the case of stable plane-wave detonation, high propagation and reaction rates combine to reduce the hydrodynamic system to a one-dimensional system which needs to be considered for only a limited length. Diameter effects in detonation are quite plainly the result of an upward displacement of the escape rate-pressure curve due to the radial components of gas flow. Those concerned with the stability of operation of solid-propellant rockets have applied these concepts quite successfully.

As applied to the growth of detonation, two or three simple cases can be visualized. First, the reaction may be initiated by an external impulse which is vigorous enough to start it at a point on the curve above (b), in which case it will grow to a stable detonation in a very short time. Second, it may start as a stable deflagration (point (a)), but a perturbation of the system, such as a change in loading density with length, may be enough to displace one or the other

curve so that they no longer intersect at (a) and (b) but only at (c). Third, it may start as stable deflagration and the discharge system progressively change (as with a lengthening discharge path) in such a manner that (a) approaches (b) until they coincide.

This last case probably explains the results described by Dr. Roth, as well as those which I described earlier, in which a deflagration grew quite suddenly to a detonation after a reasonably reproducible interval.

In all of these cases the rate of growth is determined by the evolution rate-pressure curve and the pressure-time curve. The pressure-time curve is in turn determined by the difference between the rate of evolution and the rate of escape and the free and interstitial volume of the system. In some cases, the elastic or strength properties of the confining system may also be important. Application of these concepts depends upon an extension of our knowledge of propagation rate-pressure relationships over the whole pressure spectrum.

DR. BRUNAUER: Perhaps you will forgive me, now, if I talk about the military angle of the situation. Since we in the Navy, as you so well know, are used to assigning priorities to various projects, I'd like to tell you which, in my opinion, is the most important part of the field to work on. And by that, I mean not from the theoretical point of view—because I guess from the fundamental scientific point of view any problem is as important as any other—but rather from the point of view of what is most urgent for us to know to help us in our further research.

Military explosives have three important properties, none of which are physically definable quantities: power, sensitivity, and stability. There are no units of measurement for these, either physical or chemical. About stability we need not worry. Little was said about it in this conference. That's a chemical problem, and apparently most of our interests, in this particular group, happen to be in physics. But two of the properties have been discussed here—sensitivity and power. The word "power" has not been used, but its manifestations, such as shock waves, have been discussed.

A lot of good and interesting work is in progress in the field of sensitivity, but I'd like to tell you right now that the thing which interests us most is power. I think we can tackle the problem of sensitivity. We know how to detonate explosives. We also know how to avoid detonation of explosives—namely, by careful handling.

So I don't mean to say for a moment that it would not be important for us to know a lot more about sensitivity, but I do believe that that is not the most important thing for us to find out quickly to help us in our research work. What we would like to know more about is how to produce powerful explosives. In other words, explosives which produce a lot of damage—more than our present explosives—by means of shock waves or fragmentation or shape charges.

Even within that field, permit me to specify what I consider as the most urgent for us to find out. I mentioned it, and Dr. Kirkwood mentioned it; Dr. Kistiakowsky mentioned it, and several others, without assigning priority. We so far have mentioned it along with all the other problems.

As I see it, the properties of the shock waves and the detonation velocities are not very sensitive to the form of the equation of state. I'm not sure whether our trouble at present is due to the fact that we don't have the best possible equation of state; it may be that the Kistiakowsky-Wilson equation of state would serve us almost as well as a better equation of state. Our main problem is this: that we encounter, now, explosives which have a much larger fraction of solid products than any explosives with which we dealt in World War II. Within a few weeks, I am going to recommend to the Bureau of Ordnance a new explosive and I expect it will be adopted. It's a modification of an old one, but a more powerful one, and a more damaging one for underwater purposes. It has twice as much aluminum in it as our present explosive. Our present explosive is the best military explosive used at present in the United



States. This new one will contain twice as much aluminum. In other words, there will be more solid products in the explosion.

Now, since this explosive is already developed, perhaps we don't need any further theory for it. However, we have in the books a whole series of new explosives which have been inadequately tested, or not tested at all, but which appear good on paper. All of them have the common characteristic of having more power, and a much higher percentage of solid products than those used in World War II. So our No. 1 problem is to find out how to handle the theory of these particular types of explosives. The reason why we need it so urgently is because of the shortage of manpower for experimentation.

If Dr. Brinkley could grind out for us the results, after the theoretical considerations have been settled, he could save us a lot of experimentation for which we don't have the manpower. In other words, the progress will be rather slow in the field unless theory helps us out on this score.

If a new equation of state can take care of it, of course it would be a tremendous help. But if there are other considerations necessary to find out what happens to these solids, and to take them into account in the detonation process, then I believe this is a field about which I should like to ask Kirkwood and the others to think seriously, and to help us out in the shortest possible time.

DR. SNAY: At NOL we also made theoretical investigations on explosives which form solid reaction products. A theory is now developed for such cases, and I don't think that the presence of solid particles is the reason for difficulties encountered in calculations on such explosives. The greatest difficulty we have at present is that we do not know accurately enough what the reaction products of an explosive are. For instance, it is still an open question whether the heat of explosion of TNT is 1000 cal/gram or 800 cal/gram, or somewhere between these values, corresponding to the various possible compositions of the reaction products. Our knowledge is even more scanty in the case of highly aluminized explosives. Questions like these, of course, could be attacked by means of equilibrium calculations. Although the determination of the detonation pressure and gas density (and perhaps the evaluation of the empirical parameters of an equation of state) is not very sensitive to the accuracy of the equation of state used, the equilibrium calculations are. Here, we are in particular need of a good equation of state for highly compressed gases.

We made calculations on detonation phenomena assuming the solid particles to be either compressible or incompressible. Comparison of such calculations with experiments have shown that the adiabatic compressibility of the solids can usually be neglected in comparison with that of the gases. However, the actual density of the solids at the Chapman-Jouguet point is an important factor which must be taken into account. This magnitude can be evaluated from measured detonation rates. For carbon we had the interesting result that at high loading densities the assumption of the diamond state leads to a good agreement with the experimental evidence, but the assumption of graphite does not. Perhaps this is a possible way to make diamonds.

DR. BRUNAUER: I'm sorry I'm not entirely as optimistic as Dr. Snay. Just what Dr. Snay said points up one of the difficulties. He stated that there is a considerable discrepancy, unless he assumes that the carbon is in the diamond state. But the point is that it is not a healthy situation if one has to assume such artificial things in order to explain the results where carbon is the product.

Now, as far as aluminum oxide is concerned, there are no two varieties of aluminum oxide of widely differing densities so that we can manipulate between them, and I don't know yet what the results of Dr. Snay are on that. But it seems to me that there are a lot of other things also to be considered. What happens with the solids? I mean, how are they heated up, and do they move or not move in the detonation process, and so on?

DR. KISTIAKOWSKY: In the great diamond controversy, the answer, I think, is really this: that the carbon which is formed is not in a state of graphite. I mean, that's come out of our calculations on acetylene-oxygen mixtures. But in the sense of such calculations, you cannot distinguish between diamonds and what I might call an activated carbon—that is, an extremely finely divided soot with a very high surface area, particles only a few hundred Angstroms on the average. The surface-free energy will increase over-all energy content sufficiently so that this finely divided carbon cannot thermodynamically be distinguished from diamonds. It's extremely interesting that this result comes out in the case of solid explosives, because it certainly comes out of our acetylene calculations. I'm rather confident that you can't assume graphite as the product. But if you just say, well, you've got activated carbon, then of course you're a little freer than if you talk about diamonds. And I think you do get good agreement.

DR. SNAY: I would not like to say that the problem of the solid products is completely solved. I'm not so very optimistic about this. But there are probably some more difficult problems from the viewpoint of thermodynamics (for instance, the composition of the gases). But the difficulty is this: the equilibrium composition is very sensitive to the form of the equation of state and there is probably where we actually need a good equation of state.

DR. HIRSCHFELDER: I just wanted to raise the point that when one speaks about an equation of state, in the use of the covolume, he is really getting away with quite a bit. And you get away with it because your equations are particularly insensitive to the equation of state used. However, there's a second place where your equation of state comes in, and this is in your equilibrium constants. In your equilibrium constants you have to deal with it, and you really have to use activities in place of concentrations. And I think this is the place where one is really very sensitive to an equation of state parameter. What are you going to use for the activities of the individual components? And I think this might be the main excuse for further work along equation-of-state lines—to get decent equilibrium constants.

DR. BRUNAUER: First of all, I'd like to state that I stand corrected by Dr. Snay and Dr. Hirschfelder. I see that the equation of state is extremely important. So far, all the discussion was in the direction that it is not too sensitive for calculations of detonation velocities and detonation pressures. But it is true that the equilibrium constant is of vital importance in these calculations, and for that reason we do need the best possible equation of state.

DR. COOK: I don't want to sound like I'm trying to force anything, but if the Navy's willing to stoop to barnyard-type equations of state, the one we use is the one to calculate solids. I mean, we've tried it on lots of explosives in the dynamite industry. We have considered explosives that have very high solid residues—some of these 15-10 percent dynamites—for example, high aluminum compositions. This equation assumes that the covolume  $\alpha$  is the same function of  $V$  for all explosives—solids as well as liquids. And I think we have ample proof of this. If you're not too much concerned about theoretical justification of it, the fact that it will work means that we're in business as far as calculating detonation properties and as far as calculating available energy is concerned. That equation has been pretty well demonstrated in our studies of dynamites. I'm not going to tell you any details of that but simply that it does work—so well that I think it might be of interest to apply it.

I might say that, in cases where reactions are incomplete in the detonation wave, all you have to do is increase the diameter, and you'll eventually get to the calculated velocities on those explosives as well as you do on these military explosives. Sometimes you can't go to as high diameters as necessary—that is, to get the theoretical velocity.

In one explosive we figured that we'd have to have a 57-inch-diameter column or diameter explosive to get the theoretical velocity. But we do get theoretical velocities in a 9-inch sometimes, and sometimes even in a 6-inch, or less, in most explosives. And we have a number of velocity-vs.-diameter plots that show that the velocity has leveled off, so that we know that we're dealing with the hydrodynamic velocities. So I think the fact that it's already been tested

is a good justification to use it where it's impossible by other methods to put together the equation of state from the covolumes of the specific products.

In fact, I think that it will be a terrific problem when it comes to trying to put together an equation of state from the component parts, particularly for explosives with solid residues. Lack of heat data and lack of compressibilities, etc., are going to make it almost impossible. But the fact that, in our methods, there seems to be no distinction between completely gaseous explosive products and those with solid residue is, I think, complete justification to use it whenever you need it.

DR. BRUNAUER: Now, could I ask just this question of Dr. Cook, or anybody else? Have detonation velocities been calculated on explosions where there are no gases, for example, aluminum-liquid oxygen where the product is aluminum oxide? Can you tackle it, or can anybody else?

DR. COOK: I think that you'll find that a certain part of it is gaseous.

DR. BRUNAUER: What if you do not have any gas?

DR. COOK: I think you handle it the same way. I haven't gone clear down to a zero amount of gas on these calculations, but I've gone very low, and I don't see any reason why it won't work. If it's true that you have an explosive that will explode and produce a detonation without any gas in it, I'll predict that this method will work on that.

Incidentally, I'd like to say something else in regard to aluminum and in regard to a correlation of fragmentation and things of that sort. Maybe many of you aren't familiar with it, but we did obtain a straight-line correlation between detonation pressure and impulse—that is, the cavity effect in shape charges. Now, that was for a constant charge. And we also found that, if we make a sort of correlation of a detonation head, like Dr. Jacobs talked about, it is a linear correlation. That is, the pressure—or the detonation pressure, as calculated from the Chapman-Jouguet condition—times the size of that detonation head gives you a figure proportional to the impulse in the cavity effect, and things of that sort, e.g., end impulse, booster effects, etc.

I think that's a pretty complete correlation. I don't think there can be much objection to it. Now, as far as aluminum is concerned, those studies showed pretty well that aluminum just doesn't react in a detonation wave (in small diameter charges). This has been discussed by the French and by the Germans and by the Russians, and I think everyone has come to that opinion now. Unless it's very finely divided aluminum, very little of it, if any, gets into the detonation wave.

DR. KIRKWOOD: This has a bearing on the question of reactions yielding solid products. In calculating detonation velocities, when one gets too high a fraction of aluminum, it's possible that one will run into a pathological family of Hugoniot curves—of the kind discussed by Von Neumann—in which they have an envelope. One can express this in a different way, and that must always be taken into account in making such calculations.

DR. JACOBS: I wasn't going to suggest that, but possibly the aluminum acts as inert in small charges. That wouldn't be too surprising if rarefaction set in in a matter of a few microseconds. If the aluminum normally takes 10 or 20 microseconds to react, then for small charges you are really working on a partial-reaction Hugoniot, and you don't have to make it pathological; it could be normal.

DR. KIRKWOOD: Yes, although presumably, in an underwater explosion, you would get full reaction of the aluminum. Your remark had to do with a stick of small diameter, such as would quench the reaction with aluminum.



DR. JACOBS: In the case of underwater, where they have spherical symmetry and central detonations, you hold the gases under fairly high temperature for a considerable length of time, behind the detonation, so that they could still react, even though they may not react in the detonation zone.

DR. ROBERTS: The question of personnel came up as I was making a few visits before deciding on the program for the conference, and I tried to influence certain people to attend. Some of them had the opinion that they had made their contributions to the field of detonation and explosion and that it was time that the problems be turned over to younger people with fresh ideas. That stimulated my own thinking about the possibility of getting new men into the field of detonation to consider the problems and perhaps put new ideas and new talents to work.

I think the getting of new manpower into fields of interest to the military establishment can be considered as one of the responsibilities of the Office of Naval Research program. I've always looked at the program as producing two types of products: (1) the scientific papers which are published, and (2) probably more important, the scientific manpower to conduct future research and come out with new ideas for both technological and military development.

Now, I think this is all I really have to say on this subject, but I would like to hear opinions from the people who are in the educational field about how one might go about training new people. If we could influence a few of the real good graduate students into thinking about these problems, it would be worth while.

DR. LEWIS: I would like to see established at one or two, or more if possible, universities courses on the general subject of combustion including of course combustion of solid explosives. To my knowledge there is no university where such a course is being given. I believe this is the only way to get young people—new people—interested in the field.

DR. LAWRENCE: Pertinent to the subject of manpower and personnel, especially in connection with the younger men, is the problem of deferments. We have a young man working on a project which we understand is quite important, and his draft board began to breathe down the back of his neck. So following the practice of World War II, we put in a deferment for him with considerable confidence, and we seemed to be getting nowhere fast. Then he got married, and he was deferred, right off. We have two or three other young men that we need for this program who may be in the same category but who haven't said anything about getting married.

DR. BRINKLEY: I have just a comment to make in line with comments that have already been made on personnel problems and which I'll make in the light of an experience we had at the Bureau of Mines last summer. We found, by a close scrutiny of Civil Service regulations, that it's possible to avoid a lot of the red tape and to hire on a training basis young men who are in the course of their academic preparation. We did so in the case of three individuals last summer, and that experience was exceedingly gratifying. These three people were employed on fundamental research projects that contributed to their own professional preparation, and they were employed on projects that were of benefit to the Bureau of Mines. We were able, that way, to offer them slightly more money than they could have gotten in the usual type of summer employment open to undergraduates and graduates, and I think we were able to contribute to their training.

We had one young man in particular who did quite an extraordinary job in solving a difficult mathematical problem associated with the theory of combustion. He carried it to completion in the time available to him. And I think we had the by-product of having converted a young man of considerable promise from particle physics to continuum physics. That may provide an attractive way to interest promising people in the field, and I suggest that it's worthy of consideration not only by the other government laboratories represented here but by the industry.

I'd like, incidentally, to invite those in this audience who are in academic work to suggest to any very good undergraduate or graduate students that the Bureau of Mines will consider applications for employment of that sort. We wish to expand that program. We're not interested in them unless they're good, but we hope in that way to do our part in interesting more and more people in the general problems of continuum physics.

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